

Learning metal plasticity constitutive models via laboratory-based X-ray measurements

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Abstract

Data-rich mechanics has the potential to redefine constitutive material modelling, yet its advancement has been fundamentally limited by the absence of large-scale, high-fidelity experimental datasets capable of capturing complex multiaxial behaviour. Here, we introduce a complete experimental–computational framework that unifies large-scale laboratory dataset generation with recurrent neural-network modelling of metal plasticity. At the core of this framework is a novel laboratory-scale experimental platform that integrates energy dispersive X-ray diffraction — previously confined to scarce and costly synchrotron facilities — with optical digital image correlation to simultaneously measure diverse local multiaxial stress–strain responses from a single geometrically complex specimen. This approach enables the rapid generation of rich, high-dimensional datasets with unprecedented accessibility and throughput. Using these datasets, we demonstrate that a mechanics-constrained neural operator can accurately reconstruct highly complex yield surfaces, capturing phenomena such as tension–compression asymmetry and previously inaccessible multiaxial strain-hardening behaviour. By bridging advanced experimentation, high-throughput data generation, and transfer learning approaches, this work establishes a scalable pipeline for next-generation constitutive modelling and lays a foundation for data-rich material mechanics.

Introduction

Constitutive models that describe how engineering materials respond to external loads and environmental conditions are the key ingredient that allow design engineers to simulate the real-world applications of materials via finite element calculations or other related analyses. This simulation capability reduces costly trial and error design loops. However, accurate constitutive models are typically the weakest link in this design process. Developing these models from first-principles calculations is computationally prohibitive for non-linear mechanical properties such as plasticity and fracture [1-2], and decades of research in multi-scale modelling has anyway had limited success in quantitatively predicting important engineering properties such as the strain hardening and the fracture toughness of metals. Thus, for most engineering materials, constitutive models are typically generated and calibrated via experimentation (constrained by known physical considerations). The limited availability of data implies that traditional mechanical constitutive models have a few material-specific constants which are calibrated by iterative tuning to match the observations of simple mechanical tests, e.g., uni-/biaxial tension, torsion, and bending tests. Such an approach not only lacks the expressive power to capture the complex physics of many engineering materials, but also largely relies on experience-based physical assumptions. Thus, this data-poor approach is unsuitable for the generation of new constitutive models in an autonomous material/structural design discovery loop.

The phenomenological nature of constitutive modelling and associated design technologies makes it ripe for the application of modern data-driven methods that are exploiting techniques being developed in machine learning [3-5]. This has resulted in a proliferation of such methods [6-18]. The methods range from a model-free paradigm [6-8] to physics-informed and mechanics-constrained neural constitutive models [1,2,13-15]. These methods have largely been used for modelling non-linear elasticity [16,17] but also extend to more complex history dependent properties such as plasticity where additional hidden variables are known to often play a critical role [12-15, 18]. All these approaches require large amounts of measured data in terms of stress versus strain curves for a diverse set of multiaxial strain paths. Due to the scarcity of large and diverse multiaxial stress-strain datasets, much of the work-to-date has relied on synthetic data and therefore only reproduces established constitutive models. These synthetically trained models have not yet aided the development of models for new materials and their subsequent deployment in real-world applications.

Current materials testing techniques broadly fall into two categories [19]. The traditional Materials Testing protocol (MT1.0), which can be traced back nearly 500 years to the work of da Vinci, involves designing a range of specimens with nearly homogenous stress/strain states, i.e. the specimens are statically determinate (Fig. 1a). Consequently, in MT1.0 strain can be inferred from a global displacement measure and the stress from the applied load. Each test gives data for a single strain/stress path, and many tests are required to calibrate even slightly more advanced models, e.g. calibration of the Yld2000-2d metal plasticity constitutive model [20] requires 17 independent tests via this method. Recent advances in robotics have enabled automation of this protocol to some degree [21]. The more contemporary approach, referred to as Materials Testing 2.0 (MT2.0) attempts to reduce the material testing by using geometrically complex (and statically indeterminate) specimens to trigger inhomogeneous stress/strain states within a single specimen [22-26]. While many stress/strain states are present within a MT2.0 specimen, the static and kinematic indeterminacy implies that unlike in MT1.0 specimens, the measured global load/displacement cannot directly provide information on the local stress/strain distribution within the specimen. Image correlation techniques have allowed additional information to be gleaned from these spatially heterogeneous measurements.

Specifically, digital image correlation (DIC) [27] in two-dimensions and digital volume correlation (DVC) [28-30] in three-dimensions can be used to map spatial distributions of the displacement fields within these geometrically complex specimens and thereby estimate spatial distributions of strain. However, the local stress states remain unknown and inverse techniques invoking physical principles such as stress equilibrium and assumed functional forms of constitutive relations are employed to estimate the local stress fields [16, 23-24]. This inverse problem is typically highly ill-posed as the stress is not uniquely related to strain in history dependent materials such as metals (e.g. upon unloading a plastically deforming metal, multiple stress states are consistent with the same strain). The ill-posedness of this inverse problem, which involves solving the global equilibrium equations, is typically amplified by the inevitable measurement noise in the image correlation measurements and unknown minor misalignments in the specimen loading system [25, 30]. This makes calibration of even relatively simple constitutive models using such “full-field” data [31] prone to errors.

This recognition drove our vision to more directly measure the multiaxial stress versus strain curves at each material point in a geometrically complex specimen (Fig. 1b) and thereby effectively perform many MT1.0 measurements in a single test to overcome the deficiencies of the MT2.0 data approach. The ability to perform such measurements, in accessible laboratory facilities [32-34], creates a pathway for data-driven mechanics. Numerous innovations were required for the creation of this pathway. First, we democratised energy dispersive X-ray diffraction (EDXRD) measurements that to-date are restricted to synchrotrons. This was achieved using a laboratory-based X-ray source with a liquid metal anode [35] (which has a $\sim 10^4 \times$ photon flux compared to standard laboratory X-ray sources) combined with broadband collimation of the X-ray beam and high quantum efficiency detectors. These EDXRD measurements are performed simultaneously with optical three-dimensional (3D) DIC measurements to directly reconstruct both stress and strain fields within a geometrically complex specimen (Fig. 1b and Supplementary Movie S1). Thereby, a single test provides data for diverse stress versus strain paths without recourse to any global inverse problem solution. We refer to this technique as the Materials Testing 3.0 (MT3.0) protocol (Fig. 1b) which forms a key element of a pipeline that goes from a single test to the training of deep-learning metal plasticity constitutive models (Fig. 1b). This pipeline which comprises not only the development of the MT3.0 protocol, but also computational innovations for using the data, is demonstrated on an aerospace alloy Ti6Al4V (Grade 5) which for the sake of brevity we subsequently refer to as Ti90.

Development of the MT3.0 protocol and its validation

The vision of data-driven mechanics requires the ability to collect rich and accurate datasets in readily accessible settings, i.e. standard laboratories. We therefore developed the MT3.0 protocol (Supplementary S1) that directly provides a large number of multiaxial stress versus strain responses in polycrystalline metals from a single test on a geometrically complex specimen. The MT3.0 protocol (Fig. 1b) is based around performing EDXRD measurements using a high brilliance liquid anode Metaljet E1+ X-ray source that can deliver 1000W power with a 30 μm spot-size electron beam. This enables real-time diffraction measurements on macroscopic metallic specimens in a laboratory setting. The conical beam generated by this source was collimated by Montel mirrors to generate a low divergence beam ($\Delta\Phi \approx 0.4$ mrad) over an energy range of 20 keV- 60 keV with a spot size of D at a distance of $\sim 1\text{m}$ from the X-ray source. This focussed polychromatic X-ray beam was then used to map two orthogonal components of the elastic strain field from lattice parameter changes during the loading and unloading of geometrically complex plane stress specimens. The diffracted beams were

captured at a scattering take-off angle $2\theta = 10^\circ$ by two detectors (Fig. S1), each comprising HPGGe diodes of diameter 25 mm and thickness 20 mm that, when cooled to -192°C , absorbed $\sim 99.6\%$ of 100 keV photons. The EDXRD spectra of Ti90 and an additional Al7075 (Materials and Methods) measured in this setup (Fig. S2) confirm the established unit cell d -spacings as inferred in synchrotron facilities [36]. With load applied along the x_2 -direction, the detectors along with the loading rig and specimen were oriented such that the scattering q -vectors were along the x_1 , and x_2 directions (Fig. S1). Therefore, the detectors measured the in-plane elastic strains $\varepsilon_{11}^{\text{el}}$ and $\varepsilon_{22}^{\text{el}}$ averaged over the gauge volume of size D (see Supplementary S1 for details). Simultaneous 3D DIC measurements provided the spatial distributions of total strains ε_{ij} (see Supplementary S1 for details). These combined measurements at selected applied load steps constitute the MT3.0 protocol (Fig. 1b).

The definition of stress and strain is based on the notion of a representative volume element (RVE) [37, 38] over which effective properties can be defined. For a polycrystalline material that usually means that the RVE contains a sufficiently large number of grains that different RVEs behave identically when subjected to the same average deformations. The bulk of our study was performed on a Ti90 alloy (Materials and Methods) with grain sizes in the range 5 – 15 μm (Figs. 2a and 2b). Uniaxial tensile measurements (also see Supplementary S1 for measurements of the Lankford coefficient) in different directions demonstrate that this alloy is both elastically and plastically isotropic (Fig. 2c) with a yield strength $\sigma_0 = 935\text{ MPa}$ and Young's modulus $E = 105\text{ GPa}$. In these measurements in Fig. 2c the applied axial stress $\bar{\sigma} \equiv F/A$, where F is the applied axial load and A the cross-sectional area of the gauge section of the specimen, while $\bar{\varepsilon}$ is the average axial strain in the gauge section obtained from the DIC measurements (strain distributions are uniform in this gauge section; see inset of Fig. 2c).

We converged on a RVE of size $D = 300\text{ }\mu\text{m}$ which is approximately $20 \times$ the grain size. To validate this choice, we extracted elastic and plastic properties using a statically determinate uniaxial tensile specimen (Fig. 2a). The axial and transverse elastic strains ($\varepsilon_{22}^{\text{el}}$ and $\varepsilon_{11}^{\text{el}}$, respectively) measured via EDXRD and corresponding total strains (ε_{22} and ε_{11}) measured via DIC are included in Fig. 2d as a function of the applied axial stress $\bar{\sigma}$. In Fig. 2d we include the measurements averaged over the 9 locations within the gauge section (inset of Fig. 2c and Fig. S3a) along with the associated uncertainties as estimated in Supplementary S2. For applied stresses $\bar{\sigma} < \sigma_0 = 935\text{ MPa}$ the total and elastic strains are nearly identical as there is negligible plasticity. For an applied $\bar{\sigma} > \sigma_0$, the total strains ε_{22} and ε_{11} exceed the corresponding elastic strains with the difference being the plastic strain (e.g. the plastic strain $\varepsilon_{22}^{\text{pl}} = \varepsilon_{22} - \varepsilon_{22}^{\text{el}}$) which increases with increasing $\bar{\sigma}$. Two quantitative metrics can be extracted from the EDXRD and DIC measurements to confirm our choice of the RVE size and the accuracy of elastic strains extracted from the diffraction peaks: (i) a secant modulus which is defined as either $E_s \equiv \bar{\sigma}/\varepsilon_{22}^{\text{el}}$ or $E_s \equiv \bar{\sigma}/\varepsilon_{22}$ and (ii) an effective Poisson ratio equivalently defined as $\nu_{\text{eff}} \equiv -\varepsilon_{11}^{\text{el}}/\varepsilon_{22}^{\text{el}}$ or $\nu_{\text{eff}} \equiv -\varepsilon_{11}/\varepsilon_{22}$. For an applied stress $\bar{\sigma} < \sigma_0$ both the EDXRD and DIC inferred E_s and ν_{eff} are consistent with the expected elastic moduli values for Ti90, viz. Young's modulus of 105 GPa and elastic Poisson ratio 0.33 (Fig. 2e). At $\bar{\sigma} > \sigma_0$, plastic deformation implies that the DIC inferred E_s reduces with increasing $\bar{\sigma}$ while the EDXRD estimate of the modulus remains largely unaffected. Similarly, after the onset of plasticity, ν_{eff} inferred from the DIC measurements increases towards its plastic value of 0.5 while again the EDXRD estimate is unaffected by plasticity. These measurements, not only show that the RVE size is sufficient to accurately estimate the well-established effective polycrystalline properties but also that the choices of the energy peaks used in the EDXRD measurements provide accurate elastic strains in the presence of significant plastic deformation of the Ti90. The ability

of EDXRD to provide accurate estimates of elastic strains deep in the plastic regime implies that we can also recover local stress versus strain histories. To illustrate this, we calculate σ_{22} at the 9 measurement locations in the tensile specimen using the measured $\varepsilon_{22}^{\text{el}}$ and $\varepsilon_{11}^{\text{el}}$ and isotropic Hooke's law with Young's modulus $E = 105$ GPa and Poisson ratio $\nu = 0.33$ (Supplementary S1). The stress $\langle \sigma_{22} \rangle$ averaged over the 9 measurement locations versus the corresponding DIC strain $\langle \varepsilon_{22} \rangle$ is included in Fig. 2c. The excellent correspondence between local and global measurements in this statically determinate situation implies we can now use this protocol to measure local stress versus strain histories in statically indeterminate specimens and thereby generate large multi-axial datasets from a single test (see Supplementary S1 for further validation examples).

The data-driven mechanics pipeline: from single specimen measurements to constitutive models

Consider the statically indeterminate notched Ti90 specimen loaded in tension (Fig. 3a and Fig. S3b). A multi-axial stress/strain field is generated in the gauge section around the notch as indicated in Fig. 3a — we focus our measurements on this region. The measured applied tensile force F versus displacement δ across the gauge section is plotted in Fig. 3a and on this curve we indicate the 2 instants at which we will now discuss the pipeline in more detail. The 3D DIC measurements provide in-plane total strains $(\varepsilon_{11}, \varepsilon_{22}, \varepsilon_{12})$ while the EDXRD measurements provide estimates of the elastic strains $\varepsilon_{22}^{\text{el}}$ and $\varepsilon_{11}^{\text{el}}$ and thereby the stresses σ_{11} and σ_{22} . Using 16 EDXRD measurement locations in one quarter of the gauge section (exploiting specimen symmetry, see Fig. S3b), we obtained spatial distributions of all 3 in-plane stress components $(\sigma_{11}, \sigma_{22}, \sigma_{12})$ via a supervised mapping scheme that only enforces stress equilibrium (Supplementary S2). The measured stress fields in the top half of the specimen at the two loading instants marked in Fig. 3a are included in Fig. 3b along with the corresponding distributions of the total strains inferred from the DIC measurements. The unloaded state ($F = 0$) clearly shows a residual stress field induced by the spatially inhomogeneous plastic deformation of the statically indeterminate specimen. We emphasise that the stress fields plotted in Fig. 3b are inferred from direct measurements of the elastic strain fields without making any constitutive assumption of the plastic response of the Ti90.

Unlike the uniaxial tensile test specimen (Fig. 2c), each material point within this statically indeterminate specimen undergoes a different loading history providing a rich dataset of multi-axial responses from a single test. Examples of these histories at three selected points are included in Fig. 3c: while material points “b” and “c” lie on a plane of symmetry where $\sigma_{12} = 0$, the shear stress $\sigma_{12} \neq 0$ at material point “a”. The curves are shown as shaded bands to indicate the level of anticipated uncertainty in the measurements (Supplementary S2). All the measured local stress versus strain curves are shown in Fig. S4 — such a diverse set of stress versus strain histories provides a rich dataset for extracting the history dependent multi-axial responses of Ti90.

Local versus global calibration of a constitutive model

Modern schemes for constitutive model discovery (i.e., the MT2.0 protocol) use measurements of the distributions of total strain within statically indeterminate specimens along with global load measurement [16, 25] to extract constitutive behaviour. Since local stresses are not directly measured in the MT2.0 protocol, they are inferred by solving a global optimisation problem. While many variants exist for this optimisation the basic idea is as follows. A constitutive model (in the form of a neural operator [12, 17] or a traditional plasticity model [39, 40]) is

assumed. Then for an assumed set of parameters of this constitutive model, the global equations of equilibrium and compatibility are solved for the given geometry of the specimen and the spatial distributions of the displacement fields are inferred. The parameters of the constitutive model are then optimised by repeated solutions of these global equations to minimise the difference between the predicted and measured displacements. In general, this optimisation problem is ill-posed, especially for material that has a history dependent response. On the other hand, the MT3.0 protocol provides local measurements of both elastic and total strains thereby allowing estimation of local stresses. This enables direct calibration of constitutive models without solving the global equations of equilibrium and compatibility.

Since MT2.0 measurements are a subset of the MT3.0 measurements, we can selectively use the MT3.0 datasets to make a fair comparison of the fidelity of the two techniques in inferring constitutive behaviour. The global MT2.0 problem is more regularised if the constitutive model has fewer parameters. Therefore, we selected such a model armed with the knowledge that the Ti90 is elastically and plastically isotropic. Specifically, we selected the model of Lou et al. [39] that ensures an isotropic multiaxial plastic response by specifying a yield function Φ that only depends on the stress invariants. In the special case of a metal whose yield is pressure independent, Φ in terms of the 2nd and 3rd invariants of the deviatoric stress, J_2 and J_3 , respectively (Supplementary S3) is given by

$$\Phi \equiv a \left[(J_2^3 - cJ_3^2)^{\frac{1}{2}} - dJ_3 \right]^{\frac{1}{3}} - Y = 0. \quad (1)$$

Here $Y(\bar{\epsilon}_p)$ is the uniaxial tensile strength as a function of the effective plastic strain $\bar{\epsilon}_p$ while (c, d) are independent constants with a a scaling factor to ensure that Y is the uniaxial tensile strength (Supplementary S3). The uniaxial tensile curve $Y(\bar{\epsilon}_p)$ is known from a simple tensile test (Fig. 2c) and the task therefore simply reduces to determining the constants (c, d) that control the multiaxial behaviour.

First consider model extraction using the global indirect method using the MT2.0 data. We employed the established finite element method updating (FEMU) [16, 23] technique with 16 load instants along the load versus displacement curve in Fig. 3a. Details of the optimisation to determine (c, d) are provided in Supplementary S3 and involved using finite element calculations to solve the equilibrium and compatibility equations for the tensile loading of the notched specimen. The outcome of this optimisation are the parameters (c, d) and thereby the yield function Φ . To visualise this result, we plot Φ on the Π –plane (Supplementary S3) in Fig. 3d along with the traditional von-Mises yield surface as a reference (the von Mises yield surface has no J_3 dependence and therefore corresponds to Φ with $c = d = 0$). The MT2.0 data predicts that Ti90, which comprises body-centred cubic (BCC) and hexagonally close-packed (HCP) phases, deviates from the standard von-Mises plasticity with a higher compressive yield strength compared to the tensile strength due to a non-negligible J_3 dependence of Φ .

Next consider extraction of the Lou model parameters from the MT3.0 protocol. Now we have direct measurements of multiple multiaxial stress versus strain histories (Fig. 3c) and thus there is no need to solve field equations to extract local stresses. Rather, we can directly estimate the stress versus strain responses for a given set of multiaxial strain histories and an assumed set of (c, d) by solving just the Lou et al. [39] constitutive equations and without the need for global finite element solutions. The parameters (c, d) are again optimised to best fit the measured and predicted stress versus strain curves (Supplementary S3). The yield surface predicted by the MT3.0 protocol is included in Fig. 3d and again shows a strong

tension/compression asymmetry. However, the MT2.0 and MT3.0 predictions are in stark contrast: the MT3.0 protocol predicts a lower compressive strength compared to the tensile strength while the MT2.0 protocol makes the reverse prediction. To investigate the source of this discrepancy we include in Fig. 3d the yield strengths inferred from all 16 stress versus strain curves measured using the MT3.0 method (Fig. S4) with yield defined as the 0.2% offset from the initial linear elastic response (in Fig. 3d we have used the known isotropy of Ti90 to reflect the data on the three principal stress axes). The data is clustered about the uniaxial tensile direction, and thus the prediction of the yield surface is strongly dependent on the accuracy of the data in predicting the curvature of the yield surface about this tensile direction. Data along the compressive direction will be able to distinguish which inferred yield surface is accurate.

To obtain data with a large compressive component we conducted compressive loading of the notched specimens using anti-buckling guides (Supplementary S1 and Fig. S5). Using this diverse compressive loading dataset (Fig. S6) we estimated the 0.2% offset yield strengths included in Fig. 3d. This data clearly shows that the compressive yield strength of the Ti90 is less than its tensile strength. In fact, the compressive data is in excellent agreement with the predictions of the MT3.0 protocol made from pure tensile loading of the notched specimen. The local measurements of stress in the MT3.0 protocol imply that there is no ambiguity in the stresses and thus the curvature of the yield surface about the tensile direction is predicted accurately. This results in an accurate extrapolation and correct prediction that the compressive yield strength is lower than the tensile yield strength. On the other hand, the indirect MT2.0 protocol infers the stresses by solving the global field equations for the notched specimen. This indirect method to estimate the stresses, compounded by experimental noise in the DIC measurements, reduces the accuracy of the stress estimates. Even minor measurement noise results in an erroneous prediction of the yield surface curvature about the tensile direction which is then amplified when the yield surface is extrapolated to compressive loadings (see Supplementary S3 for further details).

Data-driven constitutive models

Predictions of the Lou et al. [39] model calibrated via the MT3.0 protocol for a range of multiaxial strain paths are also included in Fig. 3c. For reference purposes the corresponding von-Mises (J_2) predictions are included in these figures. Clearly, the Lou et al. [39] model captures the multiaxial measurements (Fig. 3c) to a significantly higher level of accuracy. However, while initial yield is well captured by the Lou et al. [39] model, the strain hardening response is reproduced with less fidelity. We therefore asked can a more accurate plasticity model be developed?

The Lou et al. [39] model was chosen as an expressive conventional plasticity model but with sufficiently few parameters to allow for a fair judgement of the MT2.0 protocol. However, the large multiaxial datasets obtained using the MT3.0 protocol are very substantially underutilised when used to calibrate just two Lou parameters (c, d). The limitation of the Lou et al. [39] model is clear — while it captures initial yield to a high level of accuracy, the relative simplicity of the equations that govern the evolution of the yield surface and the directionality of plastic flow is an expected source of the discrepancies in the predictions versus measurements of the strain hardening responses. This issue can be circumvented by employing recurrent neural operators (RNOs) [15], originally designed for processing sequential data, to capture the history dependent plastic behaviour. However, training of RNOs requires large multiaxial stress versus strain datasets [12, 15, 41]. Such datasets to-date have been unavailable with data restricted to tomographic observations [42, 43]. Thus, RNOs have relied on synthetic data to

demonstrate their theoretical capability, and it has remained unclear whether RNOs can be trained using what will inevitably be noisy and relatively limited measured data.

We developed a pipeline to use our MT3.0 measurements to train an RNO (Fig. 4a) for describing the multiaxial response of Ti90 (Supplementary S4). The pipeline, based on a transfer learning concept [44, 45], allows a RNO to be trained from measured data. Broadly it comprised two stages: (i) Pretraining the RNO with large synthetic datasets spanning diverse yield surfaces to learn generic physical structure of plasticity models, and (ii) finetuning with measured data corrected for thermodynamic violations and expanded by exploiting the known isotropy of the Ti90; see Fig. S7 for the overall workflow and Supplementary S4 for further details. The RNO predictions of the multiaxial responses (Fig. 3c) confirm the substantially increased fidelity in terms of capturing the measurements including the strain hardening response (see Table S1 for quantitative metrics). Using the same loss function (Materials and Methods), the RNO captures the measurements to within 4.5% (which is within the expected experimental error) while the Lou et al. [39] model trained using the MT3.0 data has an error of $\sim 13\%$ with respect to the measurements. The same procedure was also repeated for the notched specimen but with compression loads. Again, the RNO wins with a 9.5% loss while Lou et al. [39] in compression has an error of 14.6%.

To further validate the fidelity of the RNO model we also performed MT3.0 measurements on a more complex specimen [18] (Figs. 4b and S3c). The measured applied tensile load F versus axial displacement δ averaged over the gauge section is included in Fig. 4b and the corresponding MT3.0 measurements of the local stress versus strain responses at two selected material points (along with estimated uncertainties) shown in Fig. 4c; see Fig. S8 for a more complete dataset of local stress versus strain measurements. Comparisons between these measured multiaxial stress versus strain curves, the Lou et al. [39] model (calibrated using the MT3.0 tensile notched dataset) and the RNO predictions confirm the fidelities of the models (Fig. 4c). We emphasise that these measurements for the complex specimen were not part of the training data for the models. Unlike the notched specimen (Fig. 3), both the RNO and Lou et al. [39] models calibrated with the same dataset capture the responses with approximately equal fidelity (the loss is $\sim 14.6\%$ for both the RNO and Lou et al. [39] models, Table S1).

While multiaxial stress versus strain curves are direct outputs of constitutive models, the main practical use of constitutive models is in structural calculations, i.e. prediction of the stress and strain fields within an arbitrary structure subjected to arbitrary loading. The specimen in Fig. 4b is one such structure for which we have the measured ground truth in terms of the spatial distribution of stresses and strains. Moreover, measurements of this structure did not form the calibration/training for either the Lou et al. [39] or RNO model and therefore these measurements are ideal to validate structural predictions of these models. We implemented the Lou et al. [39] and RNO models within Finite Element (FE) frameworks (Supplementary S3 and S4, respectively) and performed three-dimensional FE calculations for the tensile loading of the complex specimen. Predictions of the stress distributions within the specimen at two stages of loading are included in Fig. 4d using both the Lou et al. [39] and RNO models calibrated/trained using the MT3.0 data of Fig. 3. The corresponding MT3.0 stress measurements are also included in Fig. 4d. Remarkable agreement between the measurements and predictions of both models is observed including accurate predictions of the residual stress state after complete unloading to $F = 0$. Similar comparisons between the measured and RNO predictions of the stress and strain fields in the notched specimen are included in Fig. 3b; see Fig. S9 for a comparison between measured and RNO predictions of strain fields within the complex specimen. Overall, the differences between the RNO and Lou et al. [39] model

predictions are relatively minor suggesting that the Lou et al. [39] model might suffice for structural calculations of Ti90.

Outlook

Data-driven materials discovery and modelling is limited by the availability of data. This has driven the development of robotic systems for materials synthesis and some forms of chemical characterisation [46, 47]. Similar progress in data-driven mechanical property characterisation requires high-quality experimental datasets that are typically only obtained using synchrotron facilities, which are costly and difficult to access. To address this limitation, we developed an innovative scientific instrument (Fig. S10 and Supplementary Movie S1) that scales synchrotron technology down to the laboratory environment. This innovation is founded on the concept of the “democratisation of synchrotron technology,” enabling the generation of rich, low-cost datasets required for data-driven modelling across experimental physics, chemistry, and materials engineering. As a demonstration of its capabilities, we established a new high-throughput testing protocol (MT3.0), whereby numerous multiaxial stress–strain curves can be extracted from a single tensile test on a geometrically complex specimen. Using the resulting dataset, we demonstrated an end-to-end pipeline encompassing experimental measurement, data curation, and the implementation of a transfer-learning framework to develop a neural-network-based metal plasticity model. Together, this democratised synchrotron platform and accompanying data pipeline establish a new paradigm for data-driven materials engineering.

Materials and Methods

Materials: The bulk of the study was conducted on a grade 5 Titanium/Aluminium/Vanadium Alloy (Ti90/Al6/V4) purchased from Goodfellow (<https://www.goodfellow.com>) in the form of 0.5 mm thick sheets. The different specimen configurations (Fig. S3) were electro-discharge machined (EDM) from these sheets. Additional tests to confirm the accuracy of the EDXRD spectra measurements for different alloys were performed using 2.5 mm thick Al90/Zn5.5/Mg2.5/Cu1.5/Si0.5 (Al7075) sheets also purchased from Goodfellow.

Loss function: In the MT3.0 protocol the measured stress $\sigma_{ij}^{(k,m),\text{exp}}$ and strain $\varepsilon_{ij}^{(k,m),\text{exp}}$ state at material locations $k = 1, \dots, \mathcal{N}$ in the specimen at loading states $m = 1, \dots, \mathcal{M}$ are obtained from the EDXRD and 3D DIC measurements, respectively. Then given a constitutive model, which could be either a traditional plasticity model such as the Lou et al. [39] model or the RNO model, we can predict the stress $\sigma_{ij}^{(k,m),\text{sim}}$ history at every material location (k) given the imposed strain path $\varepsilon_{ij}^{(k,m),\text{exp}}$. We can then define an error (loss) between the measurement and prediction as

$$\mathcal{R}_\sigma \equiv \frac{1}{\mathcal{N}} \sum_{k=1}^{\mathcal{N}} \frac{\left\| \sigma_{ij}^{(k),\text{exp}} - \sigma_{ij}^{(k),\text{sim}} \right\|_F}{\left\| \sigma_{ij}^{(k),\text{exp}} \right\|_F}, \quad (2)$$

where the Frobenius norm $\|\cdot\|_F$ is defined in Supplementary S3. This is the loss function used to evaluate the fidelity of the predictions of the RNO and Lou et al. [39] models in different settings including tension/compression of the notched specimen and tension of the complex specimen. The results are summarised in Table S1.

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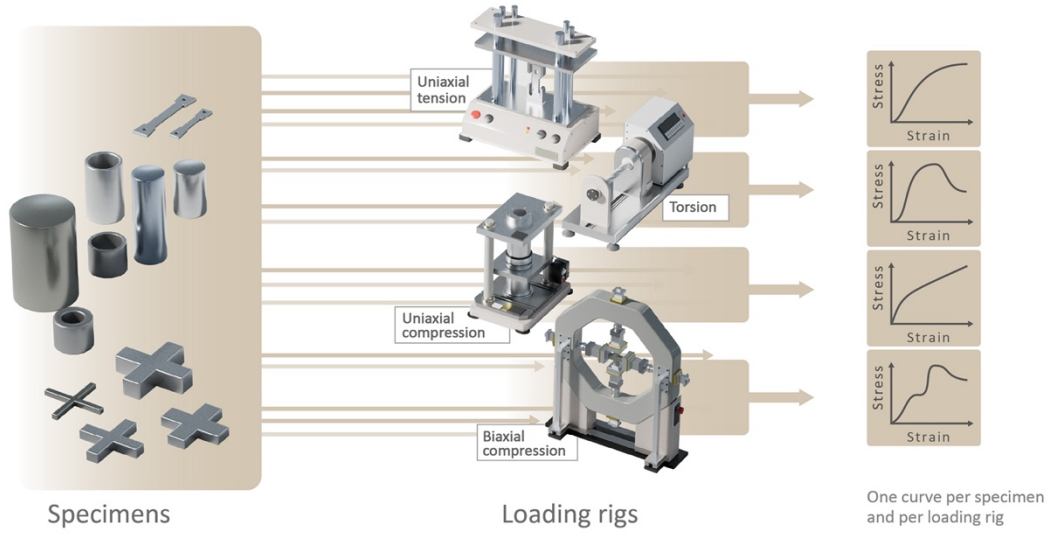
Data Availability, Code Availability

The code associated with the data curation, Lou-model calibration, recurrent neural-operator training, and figure generation is maintained on GitHub (<https://github.com/Rui696/RNO-plasticity>). Data is available on request.

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A Multiple specimens & multiple loading rigs to scan limited stress space (MT1.0)



B Single specimen & single setup to scan vast stress space (MT3.0)

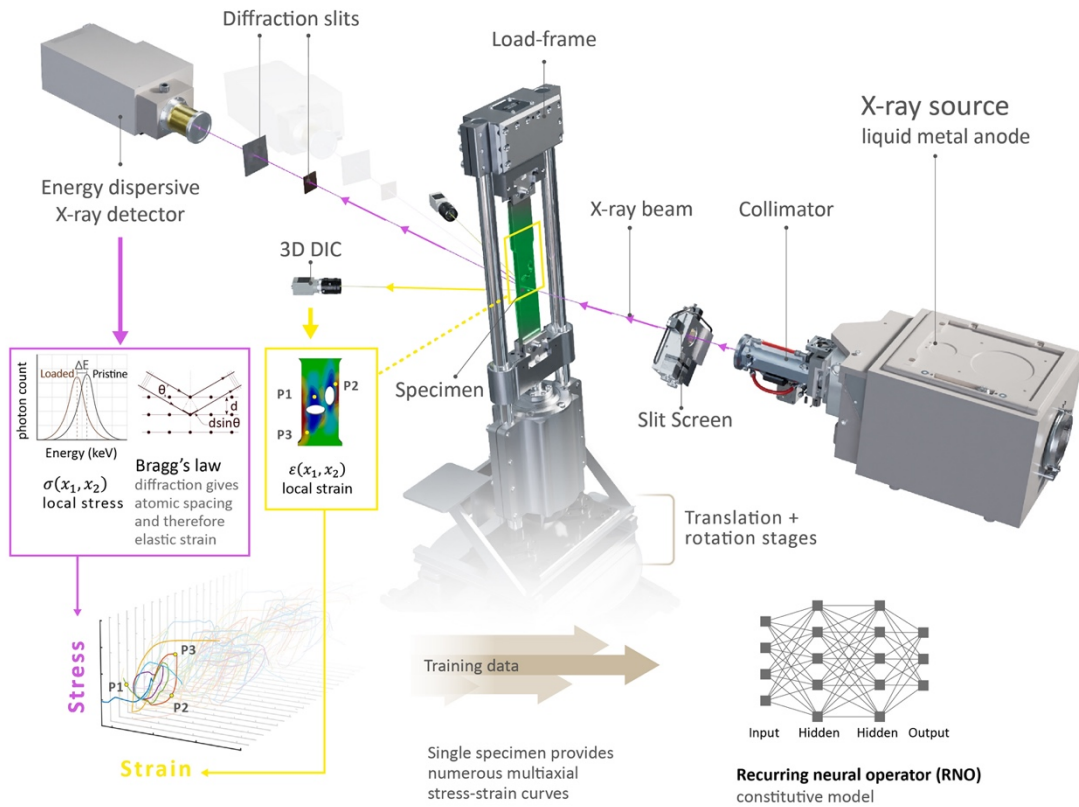


Figure 1: Evolution of materials testing into a new paradigm. Evolution of materials testing protocol from the (a) MT1.0 testing protocol which comprises numerous statically determinate specimens, each to probe a single stress state to the proposed (b) MT3.0 vision capable of making measurements of both local stresses and strains by combining X-ray diffraction with optical digital image correlation (DIC). In (b) we show a schematic (not to scale) of the laboratory MT3.0 facility developed to acquire the experimental data used to train RNO constitutive models. The facility brings energy-dispersive X-ray diffraction (EDXRD) — previously accessible only at synchrotron sources — into a standard laboratory by combining a high-flux liquid-metal-anode X-ray source, broadband Montel-mirror collimation, and two high-efficiency HPC energy-dispersive detectors for elastic-strain measurement, together with a 3D DIC system for total-strain measurement.

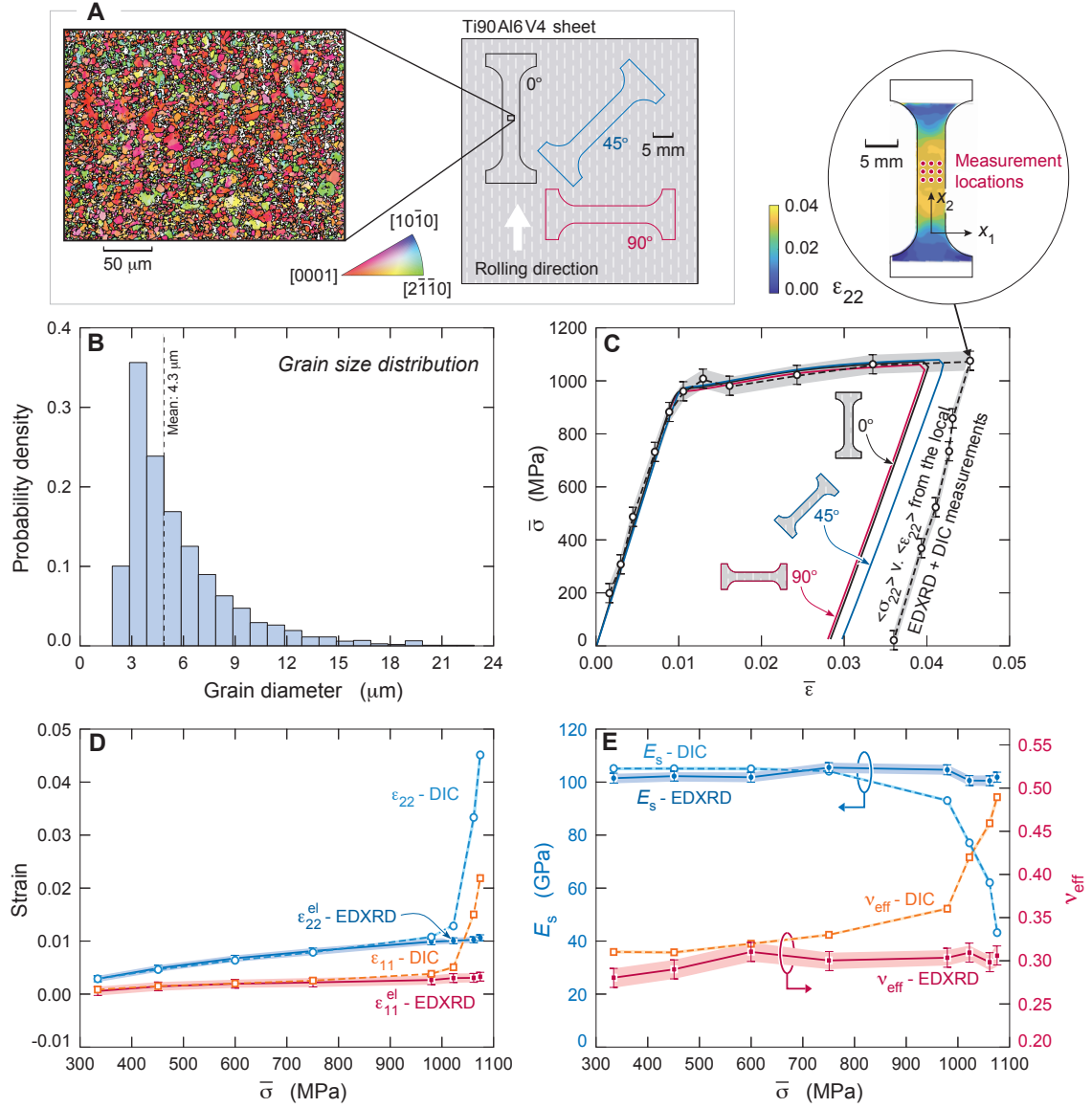


Figure 2: **Verification of the MT3.0 protocol.** (a) The Ti90 microstructure. An example electron back scattering (EBSD) image to show the grain and crystal orientation distributions and (b) the associated grain size distribution. (c) The uniaxial tensile responses ($\bar{\sigma}$ - $\bar{\epsilon}$) of the Ti90 in the 3 directions (0°, 45°, 90°) with respect to the rolling direction of the sheet as shown in (a). For the 0° -direction we also include the average $\langle \sigma_{22} \rangle$ versus $\langle \epsilon_{22} \rangle$ from the local MT3.0 measurements over 9 measurement locations in the specimen gauge section. The inset shows the locations of the 9 EDXRD measurement points along with the DIC measurement of ϵ_{22} in the 0° specimen at peak load. (d) Variation of the EDXRD axial and transverse elastic strains ($\epsilon_{22}^{\text{el}}$ and $\epsilon_{11}^{\text{el}}$) and DIC measurements of the corresponding total strains (ϵ_{22} and ϵ_{11}) as a function of the axial stress $\bar{\sigma}$. (e) The corresponding secant modulus E_s and effective Poisson ratio defined as ν_{eff} extracted from the EDXRD and DIC measurements. In (d) and (e) we show the mean over the 9 measurement locations while the error bars and shaded regions are estimates of measurement uncertainty.

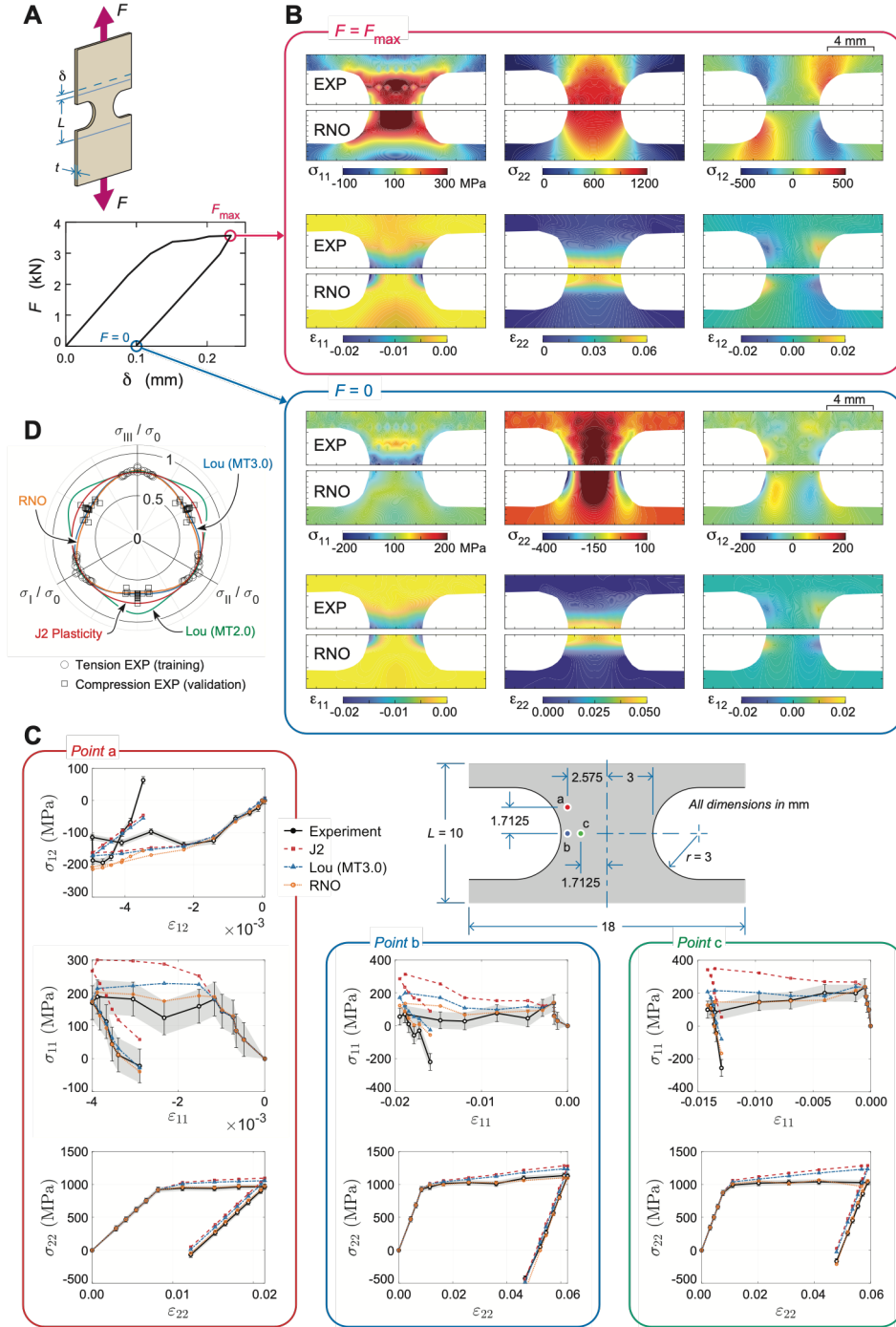


Figure 3: **Use of MT3.0 protocol to calibrate multi-axial constitutive model of Ti90 from a single test.** (a) The measured tensile force F versus axial displacement δ averaged over the gauge section of the notched specimen of thickness $t = 0.5$ mm. A sketch of the notched specimen is included with the $L = 10$ mm gauge section marked. (b) The spatial distributions of the stress and strain fields at two instants in the loading history indicated in (a). We only show the measured fields in the top half of specimen and the corresponding predictions of the RNO plasticity model in the lower half (note the anti-symmetry of the shear stress and strain fields). (c) The measured stress versus strain responses at 3 locations “a”, “b” and “c” in the gauge section as indicated in the sketch along with associated uncertainties shown by error bars and the shaded bands. We include corresponding predictions of the von-Mises plasticity model, the Lou et al. [39] model and the RNO plasticity model calibrated using only tensile MT3.0 data. (d) The predicted yield surfaces (on the Π –plane) using data from the notched specimen subjected to tensile loading. Predictions are shown by calibrating the Lou et al. [39] model using the data obtained from the MT2.0 and MT3.0 protocols as well as the RNO plasticity model trained using the MT3.0 data. The measured 0.2% offset yield stresses from the notched specimen subjected to tensile and compressive loading are also plotted.

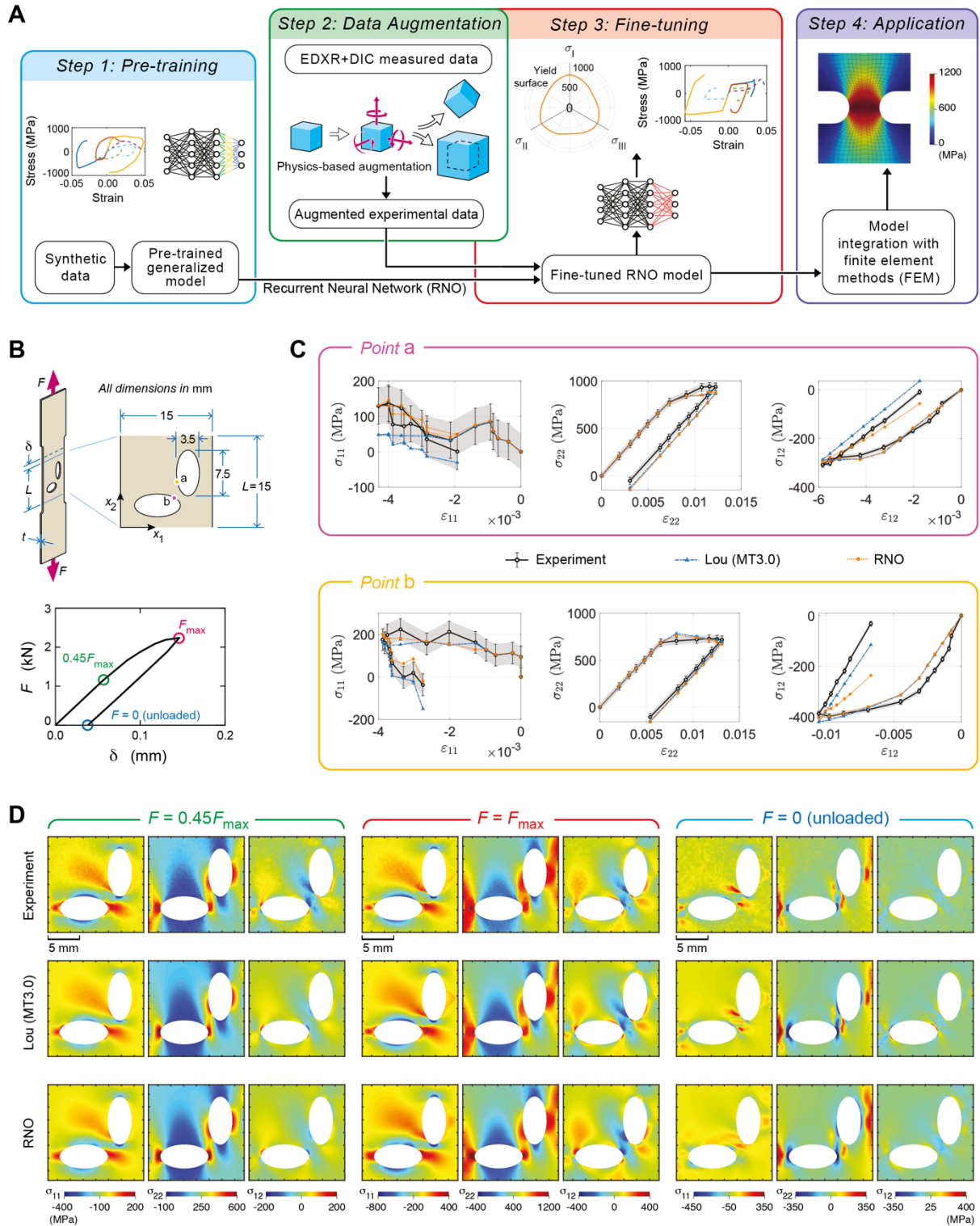


Figure 4: Training of recurrent neural operator (RNO) constitutive models and validation. (a) Sketch of the transfer learning protocol involving model pretraining and finetuning with measured data. (b) The measured tensile force F versus axial displacement δ averaged over the gauge section of the complex specimen. (c) Comparisons between the measured local stress versus strain responses at the two material locations “a” and “b” indicated in (b) along with corresponding RNO and Lou et al. [39] model (calibrated by the notched tensile MT3.0 data) predictions. The error bars and associated shaded bands indicate the measurement uncertainties. (d) Comparisons between the measured and predicted spatial distributions of the stress fields at three instants in the loading history indicated in (b). Both RNO and Lou et al. [39] model predictions trained/calibrated using the MT3.0 tensile data from the notched specimen are included.

Supplementary Information

Learning metal plasticity constitutive models via laboratory-based X-ray measurements

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Supplementary Text

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S1.2 The laboratory MT3.0 facility & experimental protocol

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S3.3 Model calibration using data obtained via the MT3.0 protocol

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S1 The MT3.0 protocol

Using geometrically complex, statically indeterminate specimens to generate a broad range of stress and strain states within a single experiment is becoming an increasingly popular strategy for accelerating the characterisation of the multiaxial response of materials [1-3]. Conventionally, the spatial distribution of strains within the specimen is measured using Digital Image Correlation (DIC) [4] or Digital Volume Correlation (DVC) [5-7], together with measurements of the global applied load. These data are subsequently used within an inverse framework to infer the underlying multiaxial constitutive response. Specifically, the local stress states are recovered by solving a global boundary value problem subject to equilibrium and compatibility constraints. This methodology, commonly referred to as MT2.0 [3], has proven highly influential; however, the associated inverse problem is often ill-posed, particularly in the presence of experimental noise [8,9].

Here, we present a fundamentally different approach in which the local stresses and strains are measured directly within a geometrically complex specimen fabricated from a crystalline solid. By eliminating the need to solve a global inverse problem, the method, which we refer to as Material Testing 3.0 (MT3.0), enables the direct experimental acquisition of numerous multiaxial stress–strain curves from a single test. This provides a robust and efficient pathway for generating high-dimensional constitutive datasets while substantially reducing the sensitivity of constitutive model extraction to measurement noise.

S1.1 The high flux laboratory X-ray source, broadband X-ray collimation and X-ray detectors

A central component of the MT3.0 protocol is the direct measurement of elastic strain through X-ray diffraction performed on macroscopic specimens in a laboratory environment. Here, we outline the key experimental and methodological requirements necessary to enable such measurements with sufficient accuracy and robustness for multiaxial constitutive characterisation.

For polycrystalline constitutive model extraction, we need to measure the “homogenised” elastic strains in polycrystalline metals defined as [10,11]

$$\varepsilon_{ij}^{\text{el}} = \frac{1}{V_{\text{RVE}}} \int_{V_{\text{RVE}}} \hat{\varepsilon}_{ij}^{\text{el}} dV, \quad (\text{S1.1})$$

where $\hat{\varepsilon}_{ij}^{\text{el}}$ is the local pointwise elastic strain within the representative volume element (RVE) and $\varepsilon_{ij}^{\text{el}}$ the average strain over the volume V_{RVE} of the RVE. This average strain $\varepsilon_{ij}^{\text{el}}$ is the strain used to define polycrystalline elastic properties such as the Young’s modulus and Poisson ratio. To measure the strain components $\varepsilon_{ij}^{\text{el}}$ we chose the energy-dispersive X-ray diffraction (EDXRD) technique. In contrast to most other X-ray diffraction techniques, EDXRD uses of a fixed scattering angle in conjunction with varying X-ray wavelength (i.e. polychromatic photons). Fixing this angle permits selection of a specific 3D measurement volume within a sample and the component of the elastic strain being measured. Moreover, because EDXRD essentially uses a “thick” Ewald sphere due to its white radiation, the whole spectrum of diffracted radiation is obtained simultaneously without rotating the sample. For these reasons the EDXRD technique is convenient for measuring the homogenised elastic strain components that feed into constitutive models.

One disadvantage of EDXRD is that to-date it has almost exclusively been only used with synchrotron radiation due to the requirement for a high flux over a wide energy range. Operating at 10W, a typical laboratory X-ray source with a nominal spot size of 30 μm has a

flux of 10^6 ph/s/mm²/mrad² [12]. On the other hand, synchrotron fluxes are $> 10^{15}$ ph/s/mm²/mrad². The low flux of laboratory X-ray sources implies that EDXRD is not a viable laboratory technique for macroscopic specimens. Recent developments [13] in laboratory X-ray sources have provided a pathway to higher fluxes. The idea is to replace the solid anode by using a liquid metal that is flowing at ~ 80 ms⁻¹. This allows using a high flux electron beam while maintaining the integrity of the anode. We utilized an Excillum MetalJet E1+ 160 kV (Excillum, Sweden) X-ray source (Supplementary Movie S1), which employs a gallium-based alloy (ExAlloy-I2 with composition by weight In 37%, Ga 47% and Sn 16%) that is maintained in a fluid state by integrated heating elements. Operating this source at an acceleration voltage of 160 kV and an emission current of 6.5 mA gives an electron beam power of 1000 W. At a nominal spot size of 30 μ m, the peak brightness is $\sim 10^{10}$ ph/s/mm²/mrad² in the 30 keV to 60 keV range representing a 10^4 increase in photon flux compared to standard laboratory sources.

The Metaljet X-ray source generates a conical beam ($\sim 20^\circ$ cone angle) but EDXRD requires parallel beam so that the diffraction angle at the fixed detector location is well-defined. This required collimation of the polychromatic conical X-ray beam which we achieved using high-precision Montel mirrors (ASTIX-f-115/885-L100-HE) manufactured by AXO Dresden. The assembly comprises two orthogonally arranged, highly polished, multilayer-coated silicon mirrors each of length 100 mm mounted in an L-shaped configuration. These multilayer coatings are fabricated to Standard Curvature Accuracy (SCA) and optimized for operation within the 20–60 keV bremsstrahlung energy range up to 1 kW. The assembly that comprises the mirror housing, motorised stages for focussing the beam, an external water-to-air cooler to maintain the temperature of the mirrors and a motorised cross-slit screen to cut-off the unwanted parts of the beam (Fig. 1b and Supplementary Movie S1). For a X-ray source spot size of 30 μ m, the resulting focal spot has a Full Width at Half Maximum (FWHM) of 300 μ m at 885 mm from the source and a beam divergence $\Delta\phi < 0.4$ mrad. Operating at the full power of 1 kW of the Metaljet source, the collimated beam had a flux of $\sim 2 \times 10^7$ ph/s.

Even with this comparatively high photon flux, at least in the context of a laboratory X-ray source, the intensity of the beam scattered from the gauge volume within the specimen remains relatively low at any given diffraction angle. To mitigate against this limitation, the EDXRD spectra are acquired using high efficiency Broad Energy Germanium Detectors (BEGe™) manufactured by Mirion Technologies (Canberra, UK). These detectors comprise a High purity Germanium (HPGe) Germanium single crystal of thickness 20 mm and diameter of 25 mm. Given the characteristics of high-purity Germanium crystals, maintaining a low temperature near liquid nitrogen levels is crucial to mitigate noise stemming from leakage current. We employed a Cryo-Pulse 5 Plus unit that is an electrically powered cryostat for use with HPGe radiation detectors and maintains the crystals at -192° C. The cryostat's high-vacuum environment ensures a clean setting for the Ge sensor and guarantees ideal working conditions for the HPGe detector, independent of external room temperatures. The 20 mm thick crystal has a photon capturing efficiency of $\sim 100\%$ for energies < 100 keV and an energy resolution of ~ 300 eV per photon. Energy calibration is performed using an Americium-241 radioactive isotope (alpha source with a decay energy of 59.54 keV). The acquisition settings utilize 1024 channels over an energy range of 0–80 keV, corresponding to an interval size of 78 eV.

In addition to detectors to measure the energy spectrum of the diffracted X-ray beams, a high spatial resolution detector to observe the direct collimated X-ray beam is also required. This detector is used to determine the position of the focal volume (gauge volume) on the specimen. We used a FDS 1.14MP high resolution imaging detector (Photonic Science, UK) with an

active imaging area $\sim 9 \text{ mm} \times 7 \text{ mm}$ and a pixel size of $6.45 \text{ }\mu\text{m}$. This allows us to locate the $D = 300 \text{ }\mu\text{m}$ collimated beam to within 2% positional accuracy.

S1.2 The laboratory MT3.0 facility & experimental protocol

Our aim is to perform combined EDXRD and DIC measurements of elastic and total strains over a geometrically complex (statically indeterminate) plane stress specimen (e.g. the notched specimen of Fig. 3a) over a number of loading stages. We shall discuss the EDXRD and DIC measurements of the elastic and total strains, respectively that form the core of the MT3.0 protocol.

EDXRD measurements: Let the collimated X-ray travel along the positive Z –direction and the X and Y – directions are in the horizontal and vertical directions (Fig. S1). The reference frame (x_1, x_2, x_3) is fixed on the specimen (Fig. S1) with the $x_1 - x_2$ in the plane of the specimen and x_3 along the specimen thickness. We wish to measure the elastic strains $\varepsilon_{11}^{\text{el}}$ and $\varepsilon_{22}^{\text{el}}$ in the x_1 and x_2 – directions, respectively. Diffraction measures the d-spacings in the direction of the q –vector and thus we need to align two q –vectors q_1 and q_2 along the x_1 and x_2 – directions, respectively. As is common in diffraction we chose a relatively small glancing angle $\theta = 5^\circ$. With the specimen lying within the plane of the load frame (Fig. S1) we rotated the load frame through $\theta = 5^\circ$ about the X –direction towards the X-ray source and also $\theta = 5^\circ$ about the Y direction. Two detectors were then located at $2\theta = 10^\circ$ with respect to the Z –direction as shown in Fig. S1. The q –vectors are set by the incoming X-ray beam and the location of the detectors such that the q –vector q_1 is aligned with the x_1 –direction with the diffraction spectrum measured by detector 1 and q –vector q_2 is aligned with the x_2 –direction with the diffraction spectrum measured by detector 2 (Fig. S1). Two pairs of pin hole slits spaced $\sim 0.5 \text{ m}$ apart guide the diffracted X-ray beam into each of the detectors (Fig. 1b). Given the spot size of the incoming X-ray beam ($300 \text{ }\mu\text{m}$) and the thickness of the plane stress specimens (0.5 mm), the gauge volume for the EDXRD measurements is a cylinder of diameter $300 \text{ }\mu\text{m}$ in the $x_1 - x_2$ in the plane of the specimen and length equal to the specimen thickness $t = 0.5 \text{ mm}$ in the x_3 –direction. Shifts in the X-ray energy spectra after loading of the specimen can then be used to infer the elastic strains $\varepsilon_{11}^{\text{el}}$ and $\varepsilon_{22}^{\text{el}}$ which will give the elastic strain averaged over this gauge volume.

DIC measurements: DIC is now a relatively standard protocol, and we performed DIC measurements using a three-dimensional (3D) stereo DIC system from Correlated Solutions to measure the spatial displacement distributions (u_1, u_2, u_3) over the face of the specimen. The optical setup utilizes two Sony IMX540 1.2" CMOS 12 MP cameras equipped with 50 mm lenses. The specimen is illuminated by a single diffused, high-intensity LED light to provide uniform, non-glare lighting over a stochastic speckle pattern. Images are processed using the Correlated Solutions Vic-3D software. The Green-Lagrange strain tensor is calculated from the displacement data using a virtual strain gauge filter size of 15. To quantify the strain measurement uncertainty, a zero-strain test was conducted by inducing a rigid body translation and rotation prior to mechanical loading. The apparent strain field inferred from this rigid body motion — which is presumably due to camera sensor noise, lighting variations, and speckle pattern quality — provides the strain uncertainty of the system to be $\sim 50 \text{ }\mu\epsilon$.

Load frame, translation stages and rotation stages: A 11 kN load frame and associated 3-axis translation stages were custom built by Psylotech (Evanston, IL, USA) for this MT3.0 facility. The load frame as well as the translation stages were motorised to enable remote control. Each stage was capable of a translation motion of $\pm 50 \text{ mm}$ with a positional accuracy of $< 1 \text{ }\mu\text{m}$. This assembly, comprising a load frame mounted on translation stages, was itself mounted on

top of a manual rotational stage and goniometer, which rotated the assembly about the Y - and X -directions, respectively (Fig. S1). This implies that the translation stages move within a rotated frame of reference aligned with (x_1, x_2, x_3) co-ordinate system. This increases the accuracy of positioning of the incoming X-ray beam on the specimen as it eliminates the need to correct for the rotation of the load frame while locating a specific location of the gauge volume on the specimen.

Measurement protocol: The MT3.0 measurements were conducted in $\mathcal{M}$ discrete loading stages as follows. The EDXRD measurements were conducted at $\mathcal{N}$ discrete gauge volume locations on the planar specimen to obtain the diffracted spectra over the gauge volume at those locations. These locations for the 3 specimens investigated in this study (the uniaxial specimen, the notched specimen and the complex validation specimen) are detailed in Fig. S3. First, both EDXRD and DIC measurements were conducted on the pristine (reference) specimen to obtain the reference state. Subsequently, the specimen was loaded to the level required for the first loading stage with the applied load measured via the load cell in the load frame. The specimen was then held with the remote displacement fixed for 2400 s to allow for relaxation. Subsequently, EDXRD spectra were acquired at each measurement location (Fig. S3) with a photon collection time of ~ 200 s per measurement location. The X-ray beam is fixed in space and diffraction spectra at each measurement location on the specimen were collected by translating the load frame in the x_1 and x_2 –directions using the motorised translation stages, which were operated in the rotated frame of reference. After the diffraction spectra were collected at all measurement locations, the load frame was then translated in the x_1 –direction to come within the view of the 3D DIC cameras and the specimen image captured to perform a DIC analysis at this loading stage. Loading was then continued and the protocol repeated for the $\mathcal{M}$ loading stages.

An important element to this protocol is positioning of the 300 μm spot of the incoming X-ray beam repeatably at the specific locations on the specimen. The incoming collimated X-ray 300 μm beam is visible on the FDS imaging detector with the photon count higher when the beam misses the specimen and lower when it is transmitted through the specimen. This allows us to accurately locate a corner of the specimen (e.g. the edge of the notch) which provides a reference point for locating specific positions of the beam on the specimen.

S1.3 The EDXRD measurements of elastic strains and the DIC measurements of total strains

While numerous complex methods [14-16] exist to extract strain/crystallographic data from diffraction measurements, we employed a relatively simple Gaussian fitting procedure to extract lattice strain data. We shall show in Section S1.4 that this simple method provides remarkably accurate estimates of the elastic strains averaged over the RVE, at least for the materials we have tested. An example EDXRD spectrum for a representative measurement location in a Ti90 specimen in its loaded and reference state is shown in Fig. S11. In this case we show the spectrum for the q_2 vector. Only the two largest peaks after a collection of 200 s are included in Fig. S11 and the associated elastic strain was extracted as follows. We first fitted a Gaussian individually to each peak. Given that the background radiation is insignificant we approximate photon count of each peak as $n(E_p) = n_0 \exp[-(-E_c)^2 / (2E_\sigma^2)]$, where E_p is the photon energy, E_c the mean energy, E_σ the standard deviation of the Gaussian and n_0 the peak amplitude of the Gaussian. This Gaussian with 3 unknowns (n_0, E_c, E_σ) was fitted to each individual peak using the built-in “fit” optimisation function in Matlab (that is part of the Curve Fitting Toolbox). Let $E_c^{(i)}$ and $E_{c0}^{(i)}$ denote the mean energies of peak (i) in the loaded and

reference state, respectively. Then the lattice strain associated with peak (i) is given by differentiating Bragg's law (at a fixed glancing angle θ) as

$$\varepsilon_{(i)}^{(q_j)} = \frac{d^{(i)} - d_0^{(i)}}{d_0^{(i)}} = -\frac{E_c^{(i)} - E_{c0}^{(i)}}{E_{c0}^{(i)}}, \quad (\text{S1.2})$$

where $d^{(i)}$ and $d_0^{(i)}$ are the d-spacings of energy peak (i) in the loaded and reference states, respectively and the superscript q_j denotes the q -vector along which this strain is measured. Let us first consider the measurement of the elastic strain $\varepsilon_{22}^{\text{el}}$ which is defined by (S1.1) as the average strain over the RVE. For the diffraction measurements this implies an average of the gauge volume and therefore given by a weighted averaged over all the energy peaks, viz.

$$\varepsilon_{22}^{\text{el}} = \frac{1}{\sum_{i=1}^n w_i} \sum_{i=1}^n w_i \varepsilon_{(i)}^{(q_2)}, \quad (\text{S1.3})$$

where a weight w_i is assigned to energy peak (i). We attempted such averaging using different choices of weights. Remarkably based on the verification results we present in Section S1.4, we observed that only using the largest energy peak (e.g. as shown in Fig. S11 for q_2 -vector) is sufficient to make a good estimate of $\varepsilon_{22}^{\text{el}}$. We interpret this to imply that the largest peak corresponds to the largest fraction of grains in the gauge volume. Thus, the shift in the largest peak provides a good estimate of the average (S1.1). A similar conclusion is also reached for the strain along q_1 , i.e. only the largest peak is also used to estimate the strain $\varepsilon_{11}^{\text{el}}$.

With the elastic strains estimated from diffraction providing an average strain over the 300 μm gauge volume, it is important that the DIC measurements of total strain use the same average volume. Although the DIC measurements have a spatial resolution of approximately 20 μm , we coarse-grained them by defining a 300 μm circular representative volume element centred on each diffraction measurement location. The total strain ε_{ij} within each circular region was then calculated as the average of all DIC measurements within that circular region. This then allowed a consistent plotting of stress versus strain curves (Figs. 3c, 4c, S4, S6, S8) where the stresses are estimated from the diffraction measurements of elastic strains and the strains from the DIC measurements.

S1.4 Validation of EDXRD measurements:

Estimation of the elastic strains via (S1.3) requires a guess on the weights w_i . We used weights such that the average (S1.3) only employed the largest diffraction peak. The validation of this choice was performed via two independent tests: (i) estimation of the known isotropic elastic properties of the Ti90 using a statically determinate specimen and (ii) global equilibrium checks using a statically indeterminate specimen.

Consider first the statically determinate tensile specimen (Fig. S3a). In this specimen the applied tensile load F directly gives the axial stress $\bar{\sigma} \equiv F/A$, where A is the cross-sectional area of the specimen in the gauge section. Then given the elastic strain measurements $\varepsilon_{11}^{\text{el}}$ and $\varepsilon_{22}^{\text{el}}$ we can estimate the isotropic elastic constants, viz. the Young's modulus E and Poisson ratio ν as

$$E \equiv \frac{\bar{\sigma}}{\varepsilon_{22}^{\text{el}}} \quad \text{and} \quad \nu \equiv -\frac{\varepsilon_{11}^{\text{el}}}{\varepsilon_{22}^{\text{el}}}. \quad (\text{S1.4})$$

These estimates are included in Fig. 2e as a function of the applied stress $\bar{\sigma}$. Not only are these estimates consistent with the known elastic moduli of Ti90 [17], they are independent of $\bar{\sigma}$

including well into the plastic domain where the total strains ε_{11} and ε_{22} exceed the corresponding elastic strains.

The above validation check confirms that the elastic strains are correctly estimated in a relatively simple statically determinate setting. Next, we demonstrate the validity of the measurements in the statically indeterminate setting of the notched specimen (Fig. S3b). The stress σ_{22} at an EDXRD measurement location is given via isotropic Hooke's law by

$$\sigma_{22} = \frac{E}{(1 - \nu^2)} [\varepsilon_{22}^{\text{el}} + \nu \varepsilon_{11}^{\text{el}}] . \quad (\text{S1.5})$$

Distributions of σ_{22} within the specimen gauge section at peak load and after unloading to zero load (Fig. 3a) are included in Fig. S12a and S12b, respectively. We now consider three horizontal lines through the gauge section as indicated. We first plot the stress σ_{22} given by (S1.5) along these lines in both Fig. S12a and S12b (the lengths of each of these lines is different and the x -axis in the plots is normalised such that $\bar{s} = 0$ at the left end of each line and $\bar{s} = 1$ at the right end). Recall the definition of traction $T_i = \sigma_{ij}n_j$, where n_j is the unit normal to the surface on which tractions are measured. Then given that the normal to the horizontal lines is $(n_1, n_2) = (0, 1)$ it follows that $T_2 = \sigma_{22}$. Now consider the average traction

$$\langle T_2 \rangle \equiv \frac{1}{\mathcal{L}} \oint_{\mathcal{L}} T_2 ds , \quad (\text{S1.6})$$

where $\mathcal{L}$ denotes the line along with the tractions are integrated. These average tractions (S1.6) are also included in Fig. S12. But force equilibrium implies that the average tractions on each of these lines can also be calculated from the known applied global force F as

$$T_{\text{global}} \equiv \frac{F}{\mathcal{L}t} , \quad (\text{S1.7})$$

where t is the specimen thickness. Since F is measured by the load cell of the load frame, a comparison between T_{global} and $\langle T_2 \rangle$ gives an independent check of the overall accuracy of the stress field σ_{22} estimated from the diffraction measurements. A comparison between T_{global} and $\langle T_2 \rangle$ in Fig. S12 shows the agreement to be within 50 MPa to 100 MPa. It is worth putting 50 MPa in context: 50 MPa corresponds to a strain of $\sim 0.05\%$ in Ti and therefore a shift of 15 eV in a diffraction energy peak located at 30 keV. This is the limit of the accuracy of the detectors and thus unsurprising that our measurements are only accurate within 50 MPa to 100 MPa.

S1.5 Estimation of isotropy and the Lankford coefficient.

The tensile stress versus strain curves (Fig. 2c) show that the Ti90 sheets are in plane isotropic. For rolled sheet metals, the Lankford coefficient (also called Lankford value, R -value, or plastic strain ratio) [18] is another measure of the plastic anisotropy. With the co-ordinate system (x_1, x_2, x_3) and uniaxial loading in the x_2 -direction $R \equiv \varepsilon_{11}^{\text{pl}}/\varepsilon_{33}^{\text{pl}}$. Isotropy implies $R = 1$. Now consider a uniaxial tension test on the statically determinate specimen (Fig. S3a). Given the applied stress $\bar{\sigma}$ and the known elastic constants (E, ν) we estimate the plastic strains as

$$\varepsilon_{22}^{\text{pl}} = \varepsilon_{22} - \frac{\bar{\sigma}}{E} , \quad \text{and} \quad \varepsilon_{11}^{\text{pl}} = \varepsilon_{11} + \nu \frac{\bar{\sigma}}{E} \quad (\text{S1.8})$$

where ε_{11} and ε_{22} are the DIC strain measurements. Then incompressibility of plastic deformation implies that $\varepsilon_{33}^{\text{pl}} = -(\varepsilon_{22}^{\text{pl}} + \varepsilon_{11}^{\text{pl}})$ enabling the R value to be estimated. The normal R_n value can then be estimated as

$$R_n \equiv \frac{1}{4}(R_0 + 2R_{45} + R_{90}) \quad (\text{S1.9})$$

where R_0 , R_{45} and R_{90} are the R values calculated from uniaxial tensile tests performed in the 0° , 45° and 90° directions with respect to the rolling direction (Fig. 2a). The R values, R_0 , R_{45} , R_{90} and R_n calculated at imposed strains $\bar{\epsilon} \sim 0.015$ and 0.025 are included in Table S2. These measurements combined with the tensile stress versus strain curves (Fig. 2c) confirm the plastic isotropy of the rolled Ti90 sheets.

S1.6 Compressive stress path measurements using anti-buckling guides

Compression experiments (Fig. 3d and S6) form a key part of the validation measurements — the stress paths in the experiments differ considerably from the tensile paths used to calibrate/train models. However, our Ti90 specimens are 0.5 mm thick with in plane dimensions ~ 20 mm. Thus, compressive loading results in buckling of the specimen in the elastic regime. To conduct compression experiments we designed anti-buckling guides (Fig. S5) which prevented the out-of-plane (in x_3 —direction) motion of the specimen while still allowing for the transmission of the collimated X-ray beam and optical observations of the gauge section for DIC measurements. The anti-buckling guide assembly comprises two steel frame halves clamped on either side of the specimen using six M3 bolts. The inner faces of the steel frames feature a precisely machined recess matching the specimen's thickness. This permitted unimpeded vertical displacement along the loading axis while strictly constraining movement in the x_3 —direction. A 5 mm thick Perspex (acrylic) window is integrated into the central cutout of each frame. This material and thickness choice is a compromise that allows continuous full-field strain mapping via the optical DIC cameras and has a relatively low X-ray attenuation while simultaneously providing sufficient stiffness to prevent the buckling of the specimen. We emphasise that friction between the Perspex and the specimen is not eliminated in this setup. However, in our MT3.0 protocol we do not use the applied global load which is influenced by friction. Rather, we directly measure local stresses and strain via EDXRD and DIC, respectively and these are direct material measurements which is not influenced by friction.

S2 Curation of experimental data

The measured EDXRD + DIC data that forms the basis of the MT3.0 protocol requires curation to be able to be used to fit both conventional (Supplementary S3) and recurrent neural operator (Supplementary S4) metal plasticity models.

S2.1 Correction for violations of Drucker's stability criteria

Uncertainties in the measurements of the elastic strain (Supplementary S2.3) can result in the data violating positive plastic dissipation as defined via the Drucker's stability criteria [19]. We shall subsequently show that such violations are relatively minor but nevertheless can create training biases for the recurrent neural operator (RNO) models (Supplementary S4). For this reason, we add a step to include minor corrections to ensure that the measured data, as used in the modelling, satisfies the Drucker's stability criteria.

Consider an elastic-plastic solid where the total strain can be additively decomposed into elastic and plastic strains. Given that elasticity is described by a linear elastic law (e.g. Hooke's law), Drucker's stability criteria reduce to a single criterion

$$d\sigma_{ij}d\epsilon_{ij}^{\text{pl}} \geq 0, \quad (\text{S2.1})$$

where $d\sigma_{ij}$ and $d\varepsilon_{ij}^{\text{pl}}$ are increments in the stress and plastic strain at any instant in the loading history. In our plane stress measurements, the non-zero components of stress are $(\sigma_{11}, \sigma_{22}, \sigma_{12})$ and thus (S2.1) is a summation of these 3 components. However, σ_{12} is not known directly from the measurements and for purposes of checking the Drucker's stability criterion we approximate (S2.1) as $(d\sigma_{11}d\varepsilon_{11}^{\text{pl}} + d\sigma_{22}d\varepsilon_{22}^{\text{pl}}) \geq 0$. This approximation is acceptable as the shear stresses σ_{12} are small compared to the direct stresses in all our measurements. Given the total and elastic strains at successive time increments t and $t + \Delta t$, the stability criterion then reduces to

$$\mathcal{S} \equiv (\Delta\varepsilon_{11}^{\text{el}} + \nu\Delta\varepsilon_{22}^{\text{el}})(\Delta\varepsilon_{11} - \Delta\varepsilon_{11}^{\text{el}}) + (\Delta\varepsilon_{22}^{\text{el}} + \nu\Delta\varepsilon_{11}^{\text{el}})(\Delta\varepsilon_{22} - \Delta\varepsilon_{22}^{\text{el}}) \geq 0, \quad (\text{S2.2})$$

where $\Delta\varepsilon_{ij}^{\text{el}} \equiv \varepsilon_{ij}^{\text{el},t+\Delta t} - \varepsilon_{ij}^{\text{el},t}$ and $\Delta\varepsilon_{ij} \equiv \varepsilon_{ij}^{t+\Delta t} - \varepsilon_{ij}^t$ are the strain increments between times $t + \Delta t$ and t . In deriving (S2.2) we have employed the fact that the total strain is decomposed into the elastic and plastic components as $\varepsilon_{ij} = \varepsilon_{ij}^{\text{el}} + \varepsilon_{ij}^{\text{pl}}$ and the plane stress isotropic Hooke's law relates stresses to elastic strains via

$$\sigma_{11} = \frac{E}{(1 - \nu^2)} [\varepsilon_{11}^{\text{el}} + \nu\varepsilon_{22}^{\text{el}}], \quad (\text{S2.3})$$

and

$$\sigma_{22} = \frac{E}{(1 - \nu^2)} [\varepsilon_{22}^{\text{el}} + \nu\varepsilon_{11}^{\text{el}}]. \quad (\text{S2.4})$$

The strains $(\varepsilon_{11}^{\text{el},t+\Delta t}, \varepsilon_{22}^{\text{el},t+\Delta t})$ are corrected to satisfy (S2.2) via the following two steps.

Step 1: Correction prior to the onset of plasticity

Using the stresses $(\sigma_{11}, \sigma_{22})$ at all times to $t + \Delta t$, we define a yield predictor as

$$\sigma_c = \max(|\sigma_{11}|, |\sigma_{22}|, |\sigma_{11} - \sigma_{22}|), \quad (\text{S2.5})$$

where the maximum is taken over all times in the loading history. If $\sigma_c < \sigma_0 = 935$ MPa for Ti90, we assumed that the deformation is elastic. At low stresses the elastic strains are small and within the measurement error¹ of the EDXRD. In this regime, the DIC measurements of strain are deemed accurate with plastic strains assumed to be negligible. Therefore, we set $\varepsilon_{ij}^{\text{el},t+\Delta t} = \varepsilon_{ij}^{t+\Delta t}$.

Step 2: Correction after the onset of plasticity

If $\sigma_c > \sigma_0 = 935$ MPa, the material could be undergoing active plastic straining. In these cases, the following procedure was employed to correct for violations of (S2.2). At a given time increment $t + \Delta t$ we calculate $\mathcal{S}$ given by (S2.2). If $\mathcal{S} \geq 0$ then no correction is applied, and the calculation proceeds to the next time increment with the current $t + \Delta t$ reset as increment t . If $\mathcal{S} < 0$ an optimum correction that minimises the change to the measured $\varepsilon_{ij}^{\text{el},t+\Delta t}$ is applied such that $\mathcal{S} = 0$. This is done as follows. Define corrected elastic strains by

$$\varepsilon_{11}^{\text{el,cor}} = \varepsilon_{11}^{\text{el},t+\Delta t} + \delta_1, \quad (\text{S2.6})$$

¹ Given an energy measurement accuracy of 15 eV and a diffraction peak at an energy of 30 keV, the accuracy of the elastic strain measured is $\sim 0.03\%$.

and

$$\varepsilon_{22}^{\text{el,cor}} = \varepsilon_{22}^{\text{el},t+\Delta t} + \delta_2, \quad (\text{S2.7})$$

where δ_1 and δ_2 are the imposed corrections. We then redefine $\Delta\varepsilon_{ij}^{\text{el}} \equiv \varepsilon_{ij}^{\text{el,cor}} - \varepsilon_{ij}^{\text{el},t}$ and minimise $\delta_1^2 + \delta_2^2$ subject to the constraint $\mathcal{S} = 0$. The strains $\varepsilon_{11}^{\text{el,cor}}$ and $\varepsilon_{22}^{\text{el,cor}}$ calculated by this optimisation procedure are then set as the elastic strains at the time increment $t + \Delta t$ and the calculation then proceeds to the next time increment.

An example of the corrections at material point “b” and “c” in the notched specimen subject to uniaxial tension (Fig. 3c) are illustrated in Fig. S13. As is evident the changes are small.

S2.2 Estimation of the shear stress

The shear stress σ_{12} is not a direct output of our EDXRD measurements as we only measure the two elastic strains $\varepsilon_{11}^{\text{el}}$ and $\varepsilon_{22}^{\text{el}}$. Here we discuss the method to estimate σ_{12} for the notched specimen (Fig. S3b). The idea is then easily generalisable to the more geometrically complex specimen of Fig. S3c. Under plane stress conditions the equilibrium equations read

$$\begin{aligned} \frac{\partial \sigma_{11}}{\partial x_1} + \frac{\partial \sigma_{12}}{\partial x_2} &= 0, \quad \text{and} \\ \frac{\partial \sigma_{12}}{\partial x_1} + \frac{\partial \sigma_{22}}{\partial x_2} &= 0, \end{aligned} \quad (\text{S2.8})$$

with traction-free boundary conditions on the “side” and “curved” surfaces and traction $T_1 = 0$ on the “top” and “bottom” surfaces of the specimen (Fig. S13). Loading is specified via the global force F in the x_2 –direction. Given this set of equilibrium equations and associated boundary conditions we learn the plane stress mapping

$$\{\sigma_{11}(y_i^{\text{XRD}}), \sigma_{22}(y_i^{\text{XRD}})\} \mapsto \{\sigma_{ij}(x_i^G)\} \quad (\text{S2.9})$$

where y_i^{XRD} are the EDXRD measurement location and x_i^G a grid of finely spaced locations in the gauge section of the specimen. For assumed smooth stress fields, the applied load F is implicit in $\{\sigma_{11}(y_i^{\text{XRD}}), \sigma_{22}(y_i^{\text{XRD}})\}$, viz. for any given line $\mathcal{L}$ along the x_1 – direction in the gauge section

$$F = t \oint_{\mathcal{L}} \sigma_{22}(y_i^{\text{XRD}}) ds, \quad (\text{S2.10})$$

where t is the specimen thickness. Thus, the mapping (S2.9) can be learnt without explicitly specifying F .

We implemented a supervised DenseNet Multi-Layer Perceptron (MLP) model to learn the mapping (S2.9) for the notched specimen. Our MLP borrows the dense-connectivity idea [20] where for each layer, the feature-maps of all preceding layers are used as inputs, and its own feature-maps are used as inputs into all subsequent layers. Such DenseNets alleviate the vanishing-gradient problem, strengthen feature propagation, encourage feature reuse, and substantially reduce the number of parameters. Our DenseNet MLP maps the two stresses $\{\sigma_{11}(y_i^{\text{XRD}}), \sigma_{22}(y_i^{\text{XRD}})\}$ at the N_{XRD} measurement locations y_i^{XRD} to the three stresses $\{\sigma_{11}(x_i^G), \sigma_{22}(x_i^G), \sigma_{12}(x_i^G)\}$ at the N_G locations x_i^G in the specimen gauge section. We used 5 fully connected layers of width $\mathcal{W} = 1000$ with SELU nonlinearity, such that the dimensionality of vectors passing through the MLP is $2N_{\text{XRD}} \rightarrow \mathcal{W} \rightarrow \mathcal{W} \rightarrow \mathcal{W} \rightarrow \mathcal{W} \rightarrow 3N_G$. This architecture is nonlinear and does not require a structured grid, making it well-suited for the irregular point distributions of the EDXRD sampling layouts. However, it is worth noting

that there were no equilibrium equations, weak-form residuals, or boundary condition enforcement incorporated in this model. Hence, the model must be trained on a fixed grid and does not generalize to new meshes or permutations of locations of the stresses.

Training is fully supervised using FEM-generated stress fields in the notched specimen (Fig. S3b). Thus, by construction, the training dataset automatically satisfies the 2D equilibrium equation and boundary conditions for the notched specimen employed in the experiments. The input to the MLP were the stress $(\sigma_{11}, \sigma_{22})$ at the $N_{\text{XRD}} = 16$ measurement locations in the experiments (Fig. S3b) while the output of the MLP were the three stresses $(\sigma_{11}, \sigma_{22}, \sigma_{12})$ on a grid comprising $N_G = 179$ locations in the form of an approximately uniform square grid with the locations spaced ~ 0.5 mm apart. Training was performed using FEM-generated training samples spanning 120 materials obeying the Lou et al. [21] model (Supplementary S3) with parameter $b = 0$ as well as purely elastic behaviour. We selected 120 combinations of the parameters (c, d) parameters that lie within the convex hull (Fig. S14) with the elastic and strain hardening properties listed in Table S3. For each of the 120 materials, 50 load steps were used to give the total of $K = 6000$ training datasets. Training of the MLP was performed to minimise the error defined as

$$\mathcal{E} = \frac{1}{K} \sum_{k=1}^K \frac{\|\hat{\sigma}_{ij}^{(k)} - \sigma_{ij}^{(k)}\|_F}{\|\sigma_{ij}^{(k)}\|_F}, \quad (\text{S2.11})$$

Here $\|\cdot\|_2$ is the Frobenius norm given by

$$\|\mathbf{x}_{ij}^{(k)}\|_F = \sqrt{\sum_{n=1}^{N_G} \sum_{m=1}^3 [\mathbf{x}_m^{(k,n)}]^2}. \quad (\text{S2.12})$$

In (S2.12), $\mathbf{x}_{ij}^{(k,n)}$ represents $\hat{\sigma}_{ij}^{(k,n)}$ which is the output of the MLP and/or the corresponding stress $\sigma_{ij}^{(k,n)}$ from the FEM calculation for the dataset k at location $n = 1, \dots, N_G$. Further, in (S2.12) we have employed the Voigt notation for the symmetric stress tensor such that $\sigma_{11} \rightarrow \sigma_1$, $\sigma_{22} \rightarrow \sigma_2$ and $\sigma_{12} = \sigma_{21} \rightarrow \sigma_3$. The optimised MLP had a training error $\mathcal{E} < 1.5\%$ and additional validation calculations performed using another 1200 FEM generated datasets also had errors $< 1.5\%$ confirming the fidelity of the model.

This MLP was then used on the experimental datasets (corrected to satisfy the Drucker stability criteria). An example of the MLP output fields for the notched specimen at maximum load is shown in Fig. S15. The contour plots of the measured stress fields in Fig. 3 are generated using this trained MLP. A similar procedure was also followed to train an MLP for the geometrically complex specimen (Fig. S3c) and the measured fields in Fig. 4d are outputs of this MLP. Finally, we note that the shear stresses at the EDXRD measurement locations are one of the outputs of the MLP and these shear stresses combined with the DIC measured strains are used to plot the corresponding shear stress versus shear strain histories in Figs. 3c and 4c.

S2.3 Uncertainty quantification

It is important to quantify the anticipated errors in the measured stress versus strain curves (Figs. 2 to 4 as well as Figs. S4, S6 and S8) which are used to calibrate/train and validate both the conventional Lou et al. [21] model as well as the RNO model. These curves are obtained by two separate measurements: (i) the DIC measurements give the strains and (ii) the EDXRD measurements give stresses. The total strains vary between 3% to 4% while the anticipated noise/error in the DIC measurements is $50 \mu\epsilon = 5 \times 10^{-3} \%$ (Supplementary S1). Thus, compared to the total imposed strains, the errors in the DIC measurements are negligible with

the major contribution to the errors in the measured stress versus strain curves arising from the EDXRD measurements. We focus on estimating these errors.

Given the Hooke's law relations (S2.3) and (S2.4) it follows that errors in the measured elastic strains propagate to the estimated stresses. Assuming that the uncertainty in the EDXRD measurements results in a Gaussian distribution of elastic strains, we can estimate the uncertainty of the stresses in terms of the standard deviation of the measured elastic strains. Let measurement errors result in standard deviations $S_{\varepsilon_{11}^{\text{el}}}$ and $S_{\varepsilon_{22}^{\text{el}}}$ in the measured elastic strains $\varepsilon_{11}^{\text{el}}$ and $\varepsilon_{22}^{\text{el}}$, respectively. The corresponding standard deviation of the stresses σ_{11} and σ_{22} then follow from (S2.3) and (S2.4) and are given by

$$S_{\sigma_{11}}^2 = \frac{E}{(1 - \nu^2)} [S_{\varepsilon_{11}^{\text{el}}}^2 + \nu S_{\varepsilon_{22}^{\text{el}}}^2], \quad (\text{S2.13})$$

and

$$S_{\sigma_{22}}^2 = \frac{E}{(1 - \nu^2)} [\nu S_{\varepsilon_{11}^{\text{el}}}^2 + S_{\varepsilon_{22}^{\text{el}}}^2], \quad (\text{S2.14})$$

respectively. It now remains to estimate $S_{\varepsilon_{11}^{\text{el}}}$ and $S_{\varepsilon_{22}^{\text{el}}}$.

We performed a large range of EDXRD measurements at multiple locations within the gauge section of the reference and loaded Ti90 uniaxial specimens (Fig. S3a). In both the loaded and reference cases, the stress state in the gauge section is uniform and thus the EDXRD peaks are expected to be located at the same energy at all spatial locations in a sample at a given applied global load. Recalling that we only use the highest intensity peaks in the elastic strain calculations we calculated the standard deviations of the mean energies of the highest intensity peaks for the q_1 and q_2 vectors for each loading state. These standard deviations were independent of the loading state and ~ 12 eV and ~ 8 eV along the q_1 and q_2 vectors, respectively. Given that the mean energy of the peaks in these two directions are 29.7 keV and 27.9 keV, respectively it follows that the anticipated standard deviations are $S_{\varepsilon_{11}^{\text{el}}} \approx 4.04 \times 10^{-4}$ and $S_{\varepsilon_{22}^{\text{el}}} \approx 2.86 \times 10^{-4}$. The error bars in the MT3.0 measurements (i.e. EDXRD + DIC) of σ_{11} versus ε_{11} and σ_{22} versus ε_{22} shown in Figs. 2 to 4 (as wells as Figs. S4, S6 and S8) use these standard deviations along with (S2.13) and (S2.14). Further the, estimated errors on the EDXRD strains shown in Fig. 2d are $S_{\varepsilon_{11}^{\text{el}}}$ and $S_{\varepsilon_{22}^{\text{el}}}$ given above.

The estimated errors for the modulus and Poisson ratio shown in Fig. 2e require slightly different considerations. Given the strain $\varepsilon_{22}^{\text{el}}$ or ε_{22} , the secant moduli are $E_S \equiv \bar{\sigma}/\varepsilon_{22}^{\text{el}}$ or $E_S \equiv \bar{\sigma}/\varepsilon_{22}$, where $\bar{\sigma}$ is the axial stress measured from the applied global load. Thus, E_S is non-linearly related to strains and we therefore calculate the standard deviation S_{E_S} by performing Monte-Carlo calculations assuming a Gaussian distribution of $\varepsilon_{22}^{\text{el}}$ or ε_{22} with the mean strain over the 9 measurement locations at the given $\bar{\sigma}$ and the standard deviations $S_{\varepsilon_{22}^{\text{el}}} = 2.86 \times 10^{-4}$ and $S_{\varepsilon_{22}} = 50 \mu\epsilon$. A similar Monte-Carlo procedure was also used for ν_{eff} .

Recall that the elastic shear strain is not directly measured and that the shear stress calculated via the MLP feedforward neural operator. We thus need to estimate the propagation of the measurement uncertainties $S_{\varepsilon_{11}^{\text{el}}}$ as $S_{\varepsilon_{22}^{\text{el}}}$ through the MLP network. We again do this via a Monte-Carlo procedure. At each load step where EDXRD data is acquired we have the stresses σ_{11} and σ_{22} at the $N_{\text{XRD}} = 16$ measurement locations. These strains are randomly perturbed by adding Gaussian noise with a zero mean and standard deviation $S_{\sigma_{11}}$ and $S_{\sigma_{22}}$ given by (S2.13)

and (S2.14), respectively. Then given this new perturbed $(\sigma_{11}, \sigma_{22})$ we can calculate a new distribution σ_{12} for that given load step. We performed 200 such simulations at each load step each with a different perturbed state $(\sigma_{11}, \sigma_{22})$. The standard deviation $S_{\sigma_{12}}$ can then be calculated at any given location and applied global load from these 200 states. This Monte Carlo procedure was then repeated at each load step. The errors shown in Fig. 3c as well as Figs. S4 and S6 for the $\sigma_{12} - \varepsilon_{12}$ plots were calculated using this procedure. Note that the errors $S_{\sigma_{12}}$ are much lower than the errors $S_{\sigma_{11}}$ and $S_{\sigma_{22}}$. This is related to the fact that σ_{12} calculated through the MLP is constrained by equilibrium considerations which in turn tends to reduce the propagation of errors. A similar procedure was also used to estimate the σ_{12} errors in the complex specimen using the MLP trained for that specimen. These errors are included in Fig. 4c and Fig. S8 in the $\sigma_{12} - \varepsilon_{12}$ plots.

S3 Calibration of a conventional plasticity model

The full-field data within the statically indeterminate specimens, whether it is only spatial distributions of strains (MT2.0) or both stress and strain (MT3.0) can be used to extract the plastic constitutive model. The Ti90 employed in this study is isotropic (Fig. 2c and Supplementary S1) and we thus constrain ourselves to isotropic plasticity models. We choose the yield function proposed by Lou et al. [21] that is not only very general but also well-posed in terms of convexity of the yield function over a wide range of parameters.

S3.1 The Lou et al. [21] isotropic plasticity model

The yield function proposed by Lou et al. [21] is one of the most general isotropic yield functions for metals written in terms of the three invariants of the Cauchy stress σ_{ij} . Define the deviatoric stress as $s_{ij} \equiv \sigma_{ij} - \delta_{ij}\sigma_{kk}/3$ where δ_{ij} is the Kronecker delta. The three invariants are then

$$I_1 \equiv \sigma_{kk}, \quad J_2 \equiv 0.5s_{ij}s_{ij} \quad \text{and} \quad J_3 \equiv \det(s_{ij}), \quad (\text{S3.1})$$

and the Lou et al. [21] yield surface Φ is written in terms of four non-dimensional parameters (a, b, c, d) as

$$\Phi \equiv a \left\{ bI_1 + \left[(J_2^3 - cJ_3^2)^{\frac{1}{2}} - dJ_3 \right]^{\frac{1}{3}} \right\} - Y = 0, \quad (\text{S3.2})$$

where Y sets the size of the yield surface. With Y defined as the uniaxial tensile yield strength it follows that

$$a \equiv \frac{1}{\left[b + \frac{1}{3} \left\{ (27 - 4c)^{\frac{1}{2}} - 2d \right\}^{\frac{1}{3}} \right]}. \quad (\text{S3.3})$$

The three independent parameters (b, c, d) set the pressure dependence, adjust the curvature of the yield surface and strength differential effect, respectively. While b does not influence the convexity of Φ , c and d need to be constrained to ensure the convexity of the yield function Φ — the range (c, d) values over which Φ remains convex is shown in Fig. S14; see Lou et al. [21] for the details. We assume associated flow such that the plastic strain rate is given by

$$\dot{\varepsilon}_{ij}^{\text{pl}} = \dot{\varepsilon}^{\text{pl}} \frac{\partial \Phi}{\partial \sigma_{ij}}, \quad (\text{S3.4})$$

in terms of the equivalent plastic strain rate $\dot{\varepsilon}^{\text{pl}}$. Moreover, we restrict attention to the case where the yield strength follows a hardening relation of the form

$$Y = \sigma_0 + h_\sigma (\bar{\epsilon}^{pl})^n, \quad (S3.5)$$

where σ_0 is the initial yield strength with h_σ and n defining the hardening behaviour. A fit to the measured uniaxial tensile stress versus strain curve (Fig. S16) of Ti90 gives the parameters listed in Table S3. The total strain rate $\dot{\epsilon}_{ij}$ is the sum of the elastic strain rate $\dot{\epsilon}_{ij}^{el}$ and plastic strain rate $\dot{\epsilon}_{ij}^{pl}$ such that

$$\dot{\epsilon}_{ij} = \dot{\epsilon}_{ij}^e + \dot{\epsilon}_{ij}^{pl}. \quad (S3.6)$$

Elasticity is taken to be isotropic such that the elastic strain rate in terms of the Young's modulus E and Poisson's ratio ν is

$$\dot{\epsilon}_{ij}^{el} = \frac{1 + \nu}{E} \dot{\sigma}_{ij} - \frac{\nu}{E} \dot{\sigma}_{kk} \delta_{ij}. \quad (S3.7)$$

The isotropic elastic moduli (E, ν) for polycrystalline Ti90 are well-established and this has been confirmed by our EDXRD measurements (Fig. 2c). Thus, these are taken to be known and listed in Table S3. It is also well-established that polycrystalline metals have a negligible pressure sensitivity to their plastic response [21, 22]. Therefore, throughout we set $b = 0$ and the constitutive parameter determination then reduces to using the measurements to estimate (c, d).

For an isotropic plasticity model, it is most convenient to illustrate the yield surface Φ in principal stress space ($\sigma_I, \sigma_{II}, \sigma_{III}$). Moreover, with $b = 0$, the yield surface is independent of the hydrostatic stress $(\sigma_I + \sigma_{II} + \sigma_{III})/3$ and the shape of the yield surface is the same on all octahedral planes perpendicular to the hydrostatic axis $\sigma_I = \sigma_{II} = \sigma_{III}$. It thus suffices to show the yield surface on the octahedral plane intersecting $(\sigma_I + \sigma_{II} + \sigma_{III}) = 0$. This octahedral plane is commonly referred to as the Π -plane and the yield surfaces shown in Fig. 3d are plotted on the Π -plane.

S3.2 Model calibration using data obtained via the MT2.0 protocol

Full-field measurements of displacements (and thereby total strains) over the notched specimen (Fig. S3b) along with the associated global load can be used to estimate material parameters of the Lou et al. [21] model. This is the current state-of-art and there are numerous methods to solve the associated inverse problem for material parameter determination. These methods include, but are not limited to, virtual fields method (VFM) and finite element model updating (FEMU) [2]. Here we use FEMU to fit (c, d) using 3D DIC and the corresponding global load data.

Consider the notched tensile experiments of Fig. 3. The primary observables in the experiments are the point-wise displacement fields within the specimen along the load F versus displacement δ response of Fig. 3a. An illustration of this data is shown in Fig. S17 where we include spatial distributions of the components (u_1, u_2) of the displacement for load $F = F_{\max}$ and the unloaded state ($F = 0$) along with the measured F versus δ response. The full-field measurements of u_i were conducted at $\mathcal{M} = 16$ loading stages. Each loading stage $j = 1, \dots, \mathcal{M}$ is defined by the measured global load $F^{(j)}$ and we define a displacement residue at each loading stage j as

$$\mathcal{R}_u^{(j)} \equiv \frac{1}{\mathcal{N}} \sum_{k=1}^{\mathcal{N}} \sqrt{\frac{\sum_{i=1}^2 (u_i^{(k,j),\text{exp}} - u_i^{(k,j),\text{sim}})^2}{\sum_{i=1}^2 (u_i^{(k,j),\text{exp}})^2}}, \quad (\text{S3.8})$$

where $u_i^{(k,j),\text{exp}}$ and $u_i^{(k,j),\text{sim}}$ are the measured and simulated displacement components in the i direction at $k = 1, \dots, \mathcal{N} = 65$ material points and at loading stage j (the $\mathcal{N}$ material points were chosen to be uniformly distributed over the gauge section). The total displacement residue then follows as

$$\mathcal{R}_u \equiv \frac{1}{\mathcal{M}} \sum_{j=1}^{\mathcal{M}} \mathcal{R}_u^{(j)}. \quad (\text{S3.9})$$

The aim is to minimise $\mathcal{R}_u$ with respect to the parameters (c, d) . However, we also need to ensure that Φ remains convex. The domain of (c, d) for Φ to remain convex is shown in Fig. S14 such that Φ is convex if (c, d) lies within that domain. To ensure convexity of Φ , we define a total cost function

$$\mathcal{C} \equiv \mathcal{R}_u + \mathcal{C}_p, \quad (\text{S3.10})$$

where $\mathcal{C}_p = 0$ if the values of (c, d) lie within the domain in Fig. S14 and $\mathcal{C}_p \rightarrow \infty$ if (c, d) lies outside the domain. In practise this is implemented by making $\mathcal{C}_p$ scale with the distance of (c, d) to the domain boundary when (c, d) lie outside the domain. The scaling constant is chosen such that a non-zero $\mathcal{C}_p \gg \mathcal{R}_u$. The penalty method implies that at the optimum solution $\mathcal{C}_p = 0$ and Φ is convex.

The FEMU method was implemented by coupling the package ABAQUS 2020 which performed the FE calculations to a Python script where a gradient-free Nelder-Mead [23] algorithm was implemented. The Nelder-Mead algorithm optimised the parameters (c, d) of the Lou et al. model to minimize the cost function (S3.10). The Lou et al. [21] model was implemented as a user-defined material (UMAT) in ABAQUS and the FE calculations performed using 20-node quadratic brick elements (C3D20R in the ABAQUS notation) with elements of size ~ 0.5 mm. Only the gauge section of the specimen (Fig. S3b) was analysed and the x_2 –direction displacements were constrained to zero on the bottom surface and x_2 –direction displacements $u_2 = U(t)$ applied on all the nodes on the top surface. The x_1 –direction displacements were free on both these surfaces so that the corresponding work-conjugate tractions vanish. The work conjugate to the displacement U is the applied force F . The optimisation calculations were performed as follows. Given an initial guess of the material parameters (c, d) , the displacement U was incremented until the applied load F attained the first measured load increment value $F^{(j=1)}$. At this instant, the displacement field within the specimen was determined from the FE calculations performed in ABAQUS. These displacements were exported to a Python script running the Nelder-Mead algorithm to evaluate $\mathcal{R}_u^{(j)}$ (S3.8). The FE calculations were then continued until the displacement field were determined at all the experimental load steps $j = 1, \dots, \mathcal{M}$ to then calculate $\mathcal{R}_u$ (S3.9). The Nelder-Mead algorithm then provides new guesses of the material parameters (c, d) in an attempt to minimise (S3.10). The values of the parameters at which the Nelder-Mead algorithm converges are taken as the parameters that best fit the measured data. For the MT2.0 calculations in Fig. 3c (using the tensile measurements for the notched specimen), the residue $\mathcal{C}$ at the optimal values of (c, d) is $\sim 11\%$ and the (c, d) values along with this residue are listed in Table S4. These optimised parameters were found to be insensitive to the choice of the initial guess of (c, d) .

The yield surface for Ti90 as predicted from the tensile notched specimen test using this MT2.0 protocol is plotted on the Π -plane in Fig. 3d (parameters listed in Table S4).

S3.3 Model calibration using data obtained via the MT3.0 protocol

Given measurements of local stress versus strain histories at 16 material points within the gauge section of the notched specimen (Fig. S3b), the estimation of the Lou et al. [21] model parameters is greatly simplified (and regularised). This is because the full constitutive response is available at the 16 material points without the need to solve the global boundary value problem by FE or otherwise.

Like the MT2.0 protocol we assume that the elastic properties and the uniaxial tensile strain hardening response are known (Table S3) and the tensile notched specimen data is then used to determine the Lou et al. [21] parameters (c, d) via an optimisation procedure. For each of the 16 material points we know mixed stress/strain paths. The strains $(\varepsilon_{11}, \varepsilon_{22}, \varepsilon_{12})$ are directly available from the DIC measurements, while the assumption of 2D deformation of the notched specimen implies $\varepsilon_{31} = \varepsilon_{32} = 0$. The strain ε_{33} is not known from the measurements but the notched Ti90 specimen was cut from a thin sheet (the sheet thickness $t \ll$ all leading in-plane dimensions) and thus it is reasonable to assume that plane stress conditions prevail such that $\sigma_{33} = 0$. Thus, we can solve (S3.4) to (S3.7) for a mixed loading path specified by $(\varepsilon_{11}, \varepsilon_{22}, \varepsilon_{12})$ as a function of time t and $\varepsilon_{31} = \varepsilon_{32} = \sigma_{33} = 0$ for an assumed (c, d) . Thereby we can obtain the stress versus strain histories at each of the 16 material points marked in Fig. S3b. We then define an error between the measurements and prediction as

$$\mathcal{R}_\sigma \equiv \frac{1}{\mathcal{N}} \sum_{k=1}^{\mathcal{N}} \frac{\|\sigma_{ij}^{(k),\text{exp}} - \sigma_{ij}^{(k),\text{sim}}\|_F}{\|\sigma_{ij}^{(k),\text{exp}}\|_F}, \quad (\text{S3.11})$$

where the Frobenius norm $\|\mathbf{x}_{ij}^{(k)}\|_F$ is defined analogous to (S2.12) except that the stress is now a 3D second order tensor, so the norm is given by

$$\|\mathbf{x}_{ij}^{(k)}\|_F = \sqrt{\sum_{m=1}^{\mathcal{M}} \sum_{p=1}^6 [\mathbf{x}_p^{(k,m)}]^2}. \quad (\text{S3.12})$$

In (S3.12), $\mathbf{x}_{ij}^{(k,m)}$ represents the measured and/or simulated stress components $\sigma_{ij}^{(k,m),\text{exp}}$ and $\sigma_{ij}^{(k,m),\text{sim}}$, respectively at the $k = 1, \dots, \mathcal{N} = 16$ material locations and $m = 1, \dots, \mathcal{M} = 16$ loading stages. Moreover, in (S3.12) we have employed the Voigt notation for the symmetric stress tensor such that $\sigma_{11} \rightarrow \sigma_1, \sigma_{22} \rightarrow \sigma_2, \sigma_{33} \rightarrow \sigma_3, \sigma_{23} = \sigma_{32} \rightarrow \sigma_4, \sigma_{13} = \sigma_{31} \rightarrow \sigma_5$ and $\sigma_{12} = \sigma_{21} \rightarrow \sigma_6$.

The aim is to minimise $\mathcal{R}_\sigma$ with respect to the parameters (c, d) . This is done akin to Section S3.2 where we add a penalty to (S3.11) to ensure convexity of Φ , i.e. (c, d) remains within the domain in Fig. S14. The optimisation was performed in MATLAB using the gradient-free Nelder-Mead [23] algorithm. We emphasize that these predictions are specimen independent in the sense that the algorithm does not explicitly use the notched geometry. Rather the notched geometry is only used to experimentally generate a diverse set of stress versus strain histories which are specimen independent, i.e. these histories are purely material properties. Via this optimisation we converged to $\mathcal{R}_\sigma \sim 13\%$ with the associated (c, d) parameters listed in Table S4.

The MT3.0 predictions of the Lou et al. [21] yield surface are included in Fig. 3d and consistent with the rather different (c, d) parameters obtained using the MT2.0 and MT3.0 protocols (Table S4), the yield surface shapes predicted by these two protocols are also rather different. Using the MT2.0 parameters listed in Table S4, we calculated the Lou et al. [21] predictions of the stress versus strain curves at the 16 material points on the notched tensile specimen where EDXRD data was also collected. Then using these predictions, we evaluated the error (S3.11) between the MT2.0 predictions and the measured stress versus strain curves. This gives an error $\mathcal{R}_\sigma \sim 15\%$. Similarly, using the MT3.0 parameters of the Lou et al. [21] model from Table S4 we performed FE calculations to calculate the displacement field within the tensile notched specimen and calculated the residual $\mathcal{C} = \mathcal{R}_u$ (S3.10): recall (c, d) in Table S4 lie in the convex domain of Fig. S14 and thus $\mathcal{C} = \mathcal{R}_u$. This gave an error $\mathcal{C} = 12\%$ as listed in Table S4. Thus, the residual $\mathcal{C}$ obtained via the MT2.0 and MT3.0 protocols are very similar, while $\mathcal{R}_\sigma$ is slightly lower using the MT3.0 protocol. This slightly lower $\mathcal{R}_\sigma$ gives significantly more accurate yield surface predictions as determined from the compression validation measurements shown in Fig. 3d. We proceed to discuss the reasons behind this.

S3.4 Illustration of the ill-posed nature of the MT2.0 protocol for material property extraction

The predictions of the yield surfaces by the MT2.0 and MT3.0 protocols using the data from the notched tensile specimen differ considerably (Fig. 3d). Validation experiments reported in the main text show that the MT3.0 protocol predicts the compressive strength more accurately even though the residuals for the tensile data are very similar for both protocols (Table S4). Here we discuss the reasons for the lower fidelity of the MT2.0 protocol.

Both the MT3.0 and MT2.0 protocols reproduce the strengths measured using the notched tensile specimens with comparable fidelity. This is evident in Fig. 3d, where the yield surfaces predicted by both protocols are shown alongside the yield strengths measured using the MT3.0 approach. Additional evidence is provided in Table S4, which shows that the residuals associated with the notched tensile specimens are similar for the two protocols. The discrepancies in the parameters identified using the yield function of Lou et al. [21] arise from differences in the ability of the two methods to accurately capture the local curvature of the yield surface in the vicinity of the tensile loading directions. Determining this curvature requires substantially greater accuracy than reproducing the measured strengths alone. In the MT2.0 protocol, local stresses are inferred through the solution of a global finite-element inverse problem, which limits the attainable accuracy, particularly in the presence of experimental measurement noise. As we demonstrate below, these limitations lead to significant errors in the inferred local curvature of the yield surface and, consequently, in the identified constitutive parameters.

We consider the 4 Lou et al. [21] yield surfaces with parameters (c, d) listed in Table S5 with the elastic and strain hardening properties of Ti90 (Table S3). These yield surfaces are plotted in Fig. S18a on the Π –plane and range from the circular J2 surface to 3 other shapes with different types of tension-compression asymmetry. We shall now show the ill-posedness of material property extraction using the MT2.0 protocol which only relies on displacement field measurements. We performed two sets of FE calculations to determine the spatial distributions of the displacements u_i at $\mathcal{N} = 65$ material points in the notched specimen for $\mathcal{M}$ loading stages (each loading stage was defined by the imposed global load F) for the following two cases:

- (i) FE calculations for each of the 4 choices of material properties listed in Table S5.

- (ii) A large number of randomly chosen parameters (c, d) with the only constraint that (c, d) lie within the convex domain shown in Fig. S14.

These calculations were performed as described in Section S3.2 and load F versus imposed displacement U for the choices of material parameters $m = \text{I}, \dots, \text{IV}$ are shown in Fig. S18b (note that the rather different yield surfaces of Fig. S18a all give approximately the same global force versus displacement response). Now given these 2 sets of calculations, we defined an error $\mathcal{C}_u^{(m)}$ analogous to (S3.9) for each of the cases $m = \text{I}, \text{II}, \text{III}, \text{IV}$ in Table S5 as

$$\mathcal{C}_u^{(m)} \equiv \frac{1}{\mathcal{MN}} \sum_{j=1}^{\mathcal{M}} \sum_{k=1}^{\mathcal{N}} \sqrt{\frac{\sum_{i=1}^2 \left(u_i^{(k,j),m} - u_i^{(k,j),\text{random}} \right)^2}{\sum_{i=1}^2 \left(u_i^{(k,j),m} \right)^2}}, \quad (\text{S3.13})$$

where $u_i^{(k,j),m}$ is the displacement in direction i at material point k and loading stage j for the choice of material properties m as list in Table S5. On the other hand, $u_i^{(k,j),\text{random}}$ is the corresponding displacements for one of the random choices of the material properties as mentioned in (ii) above. The error $\mathcal{C}_u^{(m)}$ gives the difference between the true displacement field for the material property choice m and another possible choice of properties. If $\mathcal{C}_u^{(m)}$ is small (e.g. within an expected experimental measurement error), then the MT2.0 protocol will be unable to distinguish between the true material properties and this other choice of material parameters.

We expect measurement errors on the order of approximately 5% in displacement. For all the random choices of (c, d) , we only selected cases where $\mathcal{C}_u^{(m)} \leq 5\%$ for each choice of m . For each choice of m , we include in Fig. S18c the true yield surface (in black) and alternative choice (in red) with an error $\mathcal{C}_u^{(m)} \sim 5\%$ — the precise value of $\mathcal{C}_u^{(m)}$ is marked. For all 4 choices of m we observe that yield surfaces very different from the true yield surface give errors $\mathcal{C}_u^{(m)} \sim 5\%$. However, in all cases the true and erroneous yield surfaces are good matches about the tensile directions where data from the notched tensile specimen primarily lies. These examples show that relatively small measurement errors can result in an erroneous prediction using the MT2.0 protocol. This issue with the MT2.0 can be better appreciated by examining the distributions of displacement within the notched specimen as predicted by the FE calculations for the true material properties and erroneous properties which give $\mathcal{C}_u^{(m)} \leq 5\%$. For this purpose, we choose the J2 yield surface ($m = \text{I}$ in Table S5) and a random choice of material parameters ($c = 1.0, d = -0.6$) which result in $\mathcal{C}_u^{(\text{I})} = 4.7\%$. Predictions of (u_1, u_2) at the two loading stages (at the peak load F_{max} and at unloaded state $F = 0$) are included in Fig. S18d for both the case of the J2 yield surface and the yield surface with $(c = 1.0, d = -0.6)$. The displacement distributions between the true and erroneous yield surface are nearly indistinguishable in all cases consistent with the low value of $\mathcal{C}_u^{(\text{I})}$. This illustrates the ill-posedness of the MT2.0 protocol in this sense that the error or residual for rather different yield surfaces can lie within the expected measurement error. This is the reason why the MT2.0 protocol resulted in a poor prediction of the Ti90 yield surface as illustrated in Fig. 3d.

S4 Training of a recurrent neural operator (RNO) plasticity model

Training of RNOs to-date has only been demonstrated using very large synthetic datasets [24, 25]. In this work we develop a protocol to train the RNO using experimental data that are both

noisy and sparse. For example, the MT3.0 dataset for the notched Ti90 tensile specimen contains only 16 multiaxial stress-strain trajectories (Fig. S4). This is orders of magnitude smaller compared to the training dataset sizes previously shown to be effective for training RNOs [24, 25]. Directly training a high-capacity, history-dependent neural operator on such a limited dataset would be prone to overfitting and poor generalization. We therefore developed a “transfer learning” pipeline (Fig. S7) that utilizes the vast available a priori knowledge of metal plasticity.

Although the experimental data for the specific Ti90 material are sparse, the broader class of elasto-plastic responses in metallic alloys is strongly constrained by substantial a priori knowledge. In particular, metal plasticity exhibits many common features, including an elastic regime, history dependence, yielding, plastic flow, and hardening. Our hypothesis is that the constitutive response of Ti90 shares a common latent representation with classical elasto-plastic constitutive models, while differing only through a relatively small number of material-specific features. Inspired by transfer-learning strategies developed in the computer-vision community [26], we exploited this prior structure through a two-stage transfer-learning framework.

In the first stage, we pretrain the RNO on a broad family of synthetic elasto-plastic constitutive models. These models are chosen to share generic physical structure, such as elastic response and hardening behaviour, while spanning diverse yield surfaces and other parameters that govern plasticity. This pretraining stage is designed to learn a transferable representation of history-dependent elasto-plastic behaviour. In the second stage, the pretrained RNO is adapted to the Ti90 MT3.0 data by fine-tuning material-specific components of the model using the available experimental trajectories. This strategy allows the network to leverage broad prior knowledge of metal plasticity while using the sparse Ti90 dataset to primarily identify the material-specific features of the material response.

The remainder of this section is structured as follows. Section S4.1 introduces the baseline RNO architecture used in this work. Sections S4.2 and S4.3 describe the proposed pretraining and fine-tuning strategies, respectively. In Section S4.4 we provide justifications for choices made in our transfer learning approach and finally in Section S4.5 we discuss the implementation of the fine-tuned RNO into a finite element framework to allow us to perform structural calculations. All relevant data and code files are available on (<https://github.com/Rui696/RNO-plasticity>).

S4.1 Summary of the RNO model and the transfer learning approach

The overall pipeline of the transfer learning protocol used to train the RNO with sparse experimental data is summarised in Fig. S7. Here we describe the approach in more detail.

S4.1.1 RNO model architecture

We briefly describe the architecture of the recurrent neural operator (RNO) used in this study and refer readers to Liu et al. [25] for a more detailed description. RNOs typically map strain as a function of time to stress as a function of time. However, in the current study, we are confined to rate-independent materials and therefore introduce a normalized time or history counter viz. $\zeta \in [0,1]$, where 0 represents the initial undeformed (reference) state and 1 the final state at the end of the loading trajectory. Consider the constitutive relation $\psi^\dagger: \{\varepsilon_{ij}(\zeta): 0 \leq \zeta \leq 1\} \mapsto \sigma_{ij}(\zeta)$ that links the strain ε_{ij} history to the Cauchy stress σ_{ij} with ζ denoting the loading-path parameter. Thus, the material response considered here is rate-

independent and the measured stress-strain histories are parameterized by the progression of loading, rather than by the elapsed time.

Inspired by the vast a priori understanding of metal plasticity that volumetric deformation is purely elastic, we decouple the volumetric and deviatoric stress contributions by decomposing the Cauchy stress tensor as

$$\sigma_{ij} = s_{ij} + B \varepsilon_{kk} \delta_{ij}, \quad (\text{S4.1})$$

where s_{ij} is the deviatoric stress, $B \equiv E / [3(1 - 2\nu)]$ the bulk modulus of the metal and δ_{ij} is the Kronecker delta. This decomposition then allows for a description that maps the deformation history to the deviatoric stress tensor through $\Psi: \{\varepsilon_{ij}(\zeta): 0 \leq \zeta \leq 1\} \mapsto s_{ij}(\zeta)$. To learn this mapping, we follow the neural-operator paradigm of Liu et al. [25] and the RNO architecture

$$\begin{cases} \sigma_{ij}(\zeta) = \mathcal{F}(\varepsilon_{ij}(\zeta), \xi_m(\zeta)) + B \varepsilon_{kk}(\zeta) \delta_{ij} \\ \dot{\xi}_m(\zeta) = \mathcal{G}(\varepsilon_{ij}(\zeta), \xi_m(\zeta)), \end{cases} \quad (\text{S4.2})$$

where $\mathcal{F}: \mathbb{R}_{\text{sym}}^{3 \times 3} \times \mathbb{R}^{d_\xi} \mapsto \mathbb{R}_{\text{sym}}^{3 \times 3}$, $\mathcal{G}: \mathbb{R}_{\text{sym}}^{3 \times 3} \times \mathbb{R}^{d_\xi} \mapsto \mathbb{R}^{d_\xi}$ with $\mathcal{F}$ and $\mathcal{G}$ selected to be Lipschitz neural operators [27], in order to suppress sensitivity of the RNO to measurement perturbations. In (S4.2), ξ_m is a d_ξ dimensional vector of internal variables that encapsulates the loading history (analogous to e.g., back-stress or accumulated plastic strain in classical plasticity theories). These internal variables are not defined *a priori* and instead they are identified as a part of the training process. We also assume that in the reference or undeformed state these variables take the value $\xi_m(0) = 0$.

S4.1.2 The transfer learning strategy

We now outline the transfer-learning strategy that comprises training the RNO with a rich synthetic dataset and then fine-tuning it with the limited available experimental data.

Stage 1: Pretraining with rich synthetic datasets

The pretraining data are generated synthetically from a family of K elasto-plastic constitutive models, denoted by $\Psi^\dagger = \{\psi_k^\dagger\}_{k=1}^K$. Each synthetic model $\psi_k^\dagger \in \Psi^\dagger$ is constructed within a common constitutive framework but is assigned a distinct yield surface φ_k within the family $\Psi^\dagger$. Thus, all models share the same generic ingredients of elasto-plasticity, including the elastic response, flow rule, and hardening mechanism, while differing in the shape of the yield surface. This construction allows the RNO to learn a transferable representation of history-dependent elasto-plastic behaviour across a controlled class of related constitutive laws.

For each constitutive model $\psi_k^\dagger$ in the pretraining set, we generate $n = 1, \dots, N$ synthetic stress-strain trajectories

$$\{\sigma_{ij}^{(n,k)} = \psi_k^\dagger[\varepsilon_{ij}^{(n)}]\}_{n=1}^N, \quad (\text{S4.3})$$

where $\varepsilon_{ij}^{(n)}$ denotes the imposed n^{th} strain history and $\sigma_{ij}^{(n,k)}$ is the corresponding stress history generated by the synthetic constitutive model (k). Then for each model $k = 1, \dots, K$, we construct a material specific RNO of the form

$$\begin{aligned}\hat{\sigma}_{ij}^{(n,k)}(\zeta) &= \mathcal{F}_{(k)}^{\text{RNO}} \circ h_{\mathcal{F}}^{\text{RNO}} \left(\varepsilon_{ij}^{(n)}(\zeta), \xi_m^{(n,k)}(\zeta) \right) + B \varepsilon_{pp}^{(n)}(\zeta) \delta_{ij}, \\ \dot{\xi}_m^{(n,k)}(\zeta) &= \mathcal{G}_{(k)}^{\text{RNO}} \circ h_{\mathcal{G}}^{\text{RNO}} \left(\varepsilon_{ij}^{(n)}(\zeta), \xi_m^{(n,k)}(\zeta) \right).\end{aligned}\tag{S4.4}$$

where $\dot{\xi}_m^{(n,k)}(\zeta)$ is the rate of learned internal state and B the bulk modulus that is common for all models (k). We assume that the dimension of the internal state ξ_m is fixed at d_ξ across all the constitutive models.

The mappings $h_{\mathcal{F}}^{\text{RNO}}: \mathbb{R}^{3 \times 3} \times \mathbb{R}^{d_\xi} \rightarrow \mathbb{R}^{d_F}$ & $h_{\mathcal{G}}^{\text{RNO}}: \mathbb{R}^{3 \times 3} \times \mathbb{R}^{d_\xi} \rightarrow \mathbb{R}^{d_G}$ are shared neural-network encoders that map the current strain and internal state into fixed latent representation spaces. These mappings are common to all K synthetic constitutive models. In contrast, the decoder maps $\mathcal{F}_{(k)}^{\text{RNO}}: \mathbb{R}^{d_F} \rightarrow \mathbb{R}^{3 \times 3}$ & $\mathcal{G}_{(k)}^{\text{RNO}}: \mathbb{R}^{d_G} \rightarrow \mathbb{R}^{d_\xi}$ are material-specific and therefore unique to each constitutive model $\psi_k^\dagger$. The shared encoders are intended to capture the common latent structure of elasto-plastic history dependence, while the material-specific decoders capture model-dependent features such as the yield-surface geometry and material parameters.

Given the target and RNO output stresses $\sigma_{ij}^{(n,\ell,k)}$ and $\hat{\sigma}_{ij}^{(n,\ell,k)}$, respectively at trajectory instants $\ell = 1, \dots, L$ for trajectories $n = 1, \dots, N$ of models $k = 1, \dots, K$, the multi-material pretraining loss is then defined as

$$\mathcal{L}_{\text{pre}} = \frac{1}{NK} \sum_{k=1}^K \sum_{n=1}^N \frac{\left\| \hat{\sigma}_{ij}^{(n,k)} - \sigma_{ij}^{(n,k)} \right\|_F}{\left\| \sigma_{ij}^{(n,k)} \right\|_F},\tag{S4.5}$$

where $\left\| x_{ij}^{(n,k)} \right\|_F$ is defined analogous to (S3.12) by summing over the $\ell = 1, \dots, L$ trajectory instants. We emphasize that the pretraining generates $k = 1, \dots, K$ models $\psi_k^\dagger$ that share encoders but have separate decoders. The protocol for jointly pretraining these K models is described in Section S4.2.

Stage 2: Finetuning with sparse data

Having pretrained the RNO on the synthetic model family introduced in Stage 1, we seek to approximate a target constitutive model that is not included in the pretraining set. In this work, this target model corresponds to the Ti90 material for which we have $p = 1, \dots, P$ target stress-strain histories

$$\left\{ \sigma_{ij}^{*(p)} = \psi_*^\dagger \left[\varepsilon_{ij}^{*(p)} \right] \right\}_{p=1}^P\tag{S4.6}$$

where each experimentally measured strain history $\varepsilon_{ij}^{*(p)}$ is paired with the corresponding stress history $\sigma_{ij}^{*(p)}$ obtained from the MT3.0 data. For consistency with the above notation, we denote this data as obtained from the experimental model $\psi_*^\dagger$.

To construct the target RNO approximation, we freeze the shared representation maps learned during Stage 1 and train only the target-specific decoder that gives a model $\psi_k^\dagger$ (prior to finetuning) closest to $\psi_*^\dagger$. With the notation used above, the resulting model is written as

$$\begin{aligned}\hat{\sigma}_{ij}^{*(p)}(\zeta) &= \mathcal{F}_*^{\text{RNO}} \circ h_{\mathcal{F}}^{\text{RNO}} \left(\varepsilon_{ij}^{*(p)}(\zeta), \xi_m^{*(p)}(\zeta) \right) + B \varepsilon_{pp}^{*(p)}(\zeta) \delta_{ij}, \\ \xi_m^{*(p)}(\zeta) &= \mathcal{G}_*^{\text{RNO}} \circ h_{\mathcal{G}}^{\text{RNO}} \left(\varepsilon_{ij}^{*(p)}(\zeta), \xi_m^{*(p)}(\zeta) \right),\end{aligned}\tag{S4.7}$$

where ξ_m^* has the same fixed dimension as in the pretrained model. The shared encoders in (S4.7) are the same Lipschitz neural-network encoders obtained in the pretraining stage and are kept fixed during fine-tuning. In contrast, the target-specific maps $\mathcal{F}_*^{\text{RNO}}: \mathbb{R}^{d_{\mathcal{F}}} \mapsto \mathbb{R}_{\text{sym}}^{3 \times 3}, \mathcal{G}_*^{\text{RNO}}: \mathbb{R}^{d_{\mathcal{G}}} \rightarrow \mathbb{R}^{d_{\xi}}$ are learned from the Ti90 data. In this way, the fine-tuning stage exploits the pretrained latent representation of generic elasto-plastic history dependence, while using the sparse target dataset primarily to identify the material-specific stress response and internal-state evolution.

The fine-tuning loss has the same form as the pretraining loss in (S4.5). Based on the target stresses $\sigma_{ij}^{*(p,\ell)}$ and the RNO predictions $\hat{\sigma}_{ij}^{*(p,\ell)}$ for loading histories $p = 1, \dots, P$ it is given by

$$\mathcal{L}_{FT} = \frac{1}{P} \sum_{p=1}^P \frac{\left\| \hat{\sigma}_{ij}^{*(p)} - \sigma_{ij}^{*(p)} \right\|_F}{\left\| \sigma_{ij}^{*(p)} \right\|_F}, \quad (\text{S4.8})$$

where $\left\| x_{ij}^{(p)} \right\|_F$ is defined analogous to (S3.12) by summing over the $\ell = 1, \dots, L$ trajectory instants. We emphasize that the definition (S4.8) is identical to the loss (S3.11) where the loading histories are analogous to the material locations and the trajectory instant analogous to the loading stage.

S4.2 Pretraining protocol using the rich synthetic datasets (Stage 1)

To overcome the data scarcity inherent in experimental characterisation, we generate a large bank of synthetic stress-strain trajectories from the Lou et al. [21] family of parameterised isotropic elasto-plastic models that span different tension-compressions asymmetries. Specifically, we choose the 3 Lou et al. [21] yield surfaces II, III, IV in Table S5 and Fig. S18 that considered representative for FCC, BCC and HCP crystal structures [21]. Synthetic data generated from these yield surface shapes are used to pre-train the RNO.

S4.2.1 Data generation and augmentation

In the pretraining stage, we consider $K = 3$ constitutive models with identical elastic and strain hardening behaviour but different yield surface shapes, characterised by a pressure independent Lou et al. [21] yield surface

$$\Phi_k \equiv a_k \left[(J_2^3 - c_k J_3^2)^{\frac{1}{2}} - d_k J_3 \right]^{\frac{1}{3}} - Y = 0, \quad (\text{S4.9})$$

where $k \in \{II, III, IV\}$ and $\{c_k, d_k\}$ are given in Table S5. With Y taken as the uniaxial tensile strength and thus a_k is related to (c_k, d_k) through (S3.3) with $b = 0$. All remaining material properties are kept fixed across the three pre-training constitutive models with the elastic and strain hardening properties listed in Table S3.

For each constitutive model k , we generate a set of $n = 1, \dots, N$ random strain paths. We divide the normalised path length $0 \leq \zeta \leq 1$, over which the deformation is prescribed, into $\ell = 1, \dots, N_\ell = 5$ equally spaced intervals with $\Delta\zeta = 1/N_\ell$. For each path and each location $\zeta_\ell = \ell/N_\ell$ along the path we compute the strain as

$$\varepsilon_{ij}^{\ell+1,n} = \varepsilon_{ij}^{\ell,n} + v_{ij}^{\ell+1,n} \varepsilon_{\max}^n \sqrt{\Delta\zeta} = 0, \quad (\text{S4.10})$$

where $v_{ij}^{\ell+1,n}$ is sampled from a Rademacher distribution for each strain component while $\varepsilon_{\max}^n$ is the maximum strain magnitude of path (n) that is sampled from the uniform distribution $\mathcal{U}(0.03, 0.05)$. The resulting discrete strain instants $\{\zeta_\ell, \varepsilon_{ij}^{\ell,n}\}_{\ell=1}^{N_\ell}$ are then interpolated using

Hermite interpolation to obtain a C^1 -continuous strain paths $\varepsilon_{ij}^{\ell,n}(\zeta)$, which provides smooth, randomly generated multiaxial strain histories for constitutive learning, suitable for operator learning. We emphasise that the random-walk sub-intervals $\Delta\zeta$ used to *define* the loading paths are conceptually distinct from the integration steps that we use to *train and query* the RNO. Specifically, the strain instants $\{\zeta_\ell, \varepsilon_{ij}^{\ell,n}\}_{\ell=1}^{N_\ell}$ set the path while the RNO integration step sets the temporal resolution at which the operator is sampled.

For each material model $k \in \{II, III, IV\}$ we generated $N = 3500$ stress-strain trajectories (90 % training + 10 % validation). To diversify the loading regimes encountered during pre-training and to sufficiently cover the multi-axial stress state manifold, we generate 8 canonical loading classes:

1. Arbitrary 3D smooth paths (cubic-Hermite-interpolated random walk).
2. Arbitrary 3D non-smooth path (piecewise-linear random walk).
3. Plane strain smooth paths ($\varepsilon_{33} = \varepsilon_{13} = \varepsilon_{23} = 0$).
4. Plane strain non-smooth paths ($\varepsilon_{33} = \varepsilon_{13} = \varepsilon_{23} = 0$).
5. Pseudo plane stress smooth paths where ε_{33} is estimated from an assumed elastic Poisson contraction $\varepsilon_{33} = -\nu/(1 - \nu)(\varepsilon_{11} + \varepsilon_{22})$.
6. Pseudo plane stress non-smooth paths.
7. True plane stress smooth paths (ε_{33} estimated to enforce $\sigma_{33} = 0$ for the given Φ_k).
8. True plane stress non-smooth paths.

The dataset comprising 3500 stress versus strain trajectories covers a wide range of stress states and covered the entire non-dimensional stress invariant space parameterised by the stress triaxiality $I_1/(3\sqrt{3}J_2)$ and Lode parameter $9J_3/(2\sqrt{3}J_2^3)$. However, the aim of the pretraining in our transfer learning approach is to teach the model the basic physics of metal plasticity as it pertains to the class of metallic alloys we are investigating, viz. an isotropic and pressure insensitive underlying plastic response. To specifically train the RNO for these physical characteristics we densified the dataset comprising 3500 stress versus strain trajectories using the following three augmentation operations. In the augmentation, given any one of the 3500 $\sigma_{ij} - \varepsilon_{ij}$ responses generated above, we generated additional responses via the following transformations.

(i) *Rotational augmentation*: Ti90 is both elastically and plastically isotropic and thus we are constraining ourselves to isotropic elastic-plastic models. Thus, a given a $\sigma_{ij} - \varepsilon_{ij}$ response, this implies that the material will also obey another equivalent response $\sigma'_{ij} - \varepsilon'_{ij}$ where

$$\sigma'_{ij} = R_{im}\sigma_{mn}R_{jn} \quad \& \quad \varepsilon'_{ij} = R_{im}\varepsilon_{mn}R_{jn}, \quad (\text{S4.11})$$

where R_{ij} is any orthogonal matrix such that $R_{ji} = R_{ij}^{-1}$ and $\det(R_{ij}) = 1$.

(ii) *Permutation augmentation*: Again, the isotropy of the Ti90 implies that the behaviour is only dependent on the isotropic stress and strain invariants. Thus, given a $\sigma_{ij} - \varepsilon_{ij}$ response, permuting the stress/strain components so as to keep the invariants unchanged generates an equivalent response. Then given a permutation matrix P_{ij} which has exactly one entry of 1 in each row and each column with all other entries equal to 0, we define augmented stresses and strains as

$$\sigma'_{ij} = P_{im}\sigma_{mn}P_{jn} \quad \& \quad \varepsilon'_{ij} = P_{im}\varepsilon_{mn}P_{jn}. \quad (\text{S4.12})$$

The material will then also obey the response $\sigma'_{ij} - \varepsilon'_{ij}$.

(iii) *Pressure augmentation*: The plastic response of the Ti90 is pressure insensitive and thus superposing an additional hydrostatic stress only results in elastic deformation. Therefore, given any $\sigma_{ij} - \varepsilon_{ij}$ response we can superimpose an arbitrary pressure p and define augmented stresses and strains given by

$$\sigma'_{ij} = \sigma_{ij} - \delta_{ij}p \quad \& \quad \varepsilon'_{ij} = \varepsilon_{ij} - \frac{\delta_{ij}p}{3B}. \quad (\text{S4.13})$$

The material will then also obey the response $\sigma'_{ij} - \varepsilon'_{ij}$.

The rotation and permutation augmentations were applied uniformly to a given measured trajectory $\sigma_{ij} - \varepsilon_{ij}$, i.e. R_{ij} and P_{ij} were invariant over the N_ℓ steps of the $\sigma_{ij} - \varepsilon_{ij}$ trajectory. On the other hand, in the pressure augmentation p was allowed to vary randomly (and smoothly) over the N_ℓ steps of the trajectory with the augmented volumetric strain p/B restricted to be less than $\max(\varepsilon_{11}, \varepsilon_{22}, \varepsilon_{33})$ so that the strain state remained approximately within the experimentally probed regime.

For each of the pretraining material dataset (e.g. $k = II$) we have 3500 trajectories. We then generated a total of 14000 trajectories from these 3500 trajectories as follows. We left 1750 trajectories unchanged and applied the following 7 augmentations to the remaining 1750 trajectories: (i) rotational augmentation, (ii) permutation augmentation, (iii) rotation + permutation augmentation, (iv) pressure augmentation; (v) pressure + rotation augmentation; (vi) pressure + permutation augmentation and (vii) pressure + rotation + permutation augmentation. Each of these augmentations generated 1750 trajectories and combined with the original unchanged 1750 trajectories we obtained a total of 14000 trajectories.

S4.2.2 Pretraining methodology and results

The model was trained using the Adam optimizer with a cosine annealing learning rate scheduler, using the loss function (S4.5). Training was performed with an initial learning rate of 5×10^{-5} a batch size of 70, and an internal variable vector size $d_\xi = 5^2$ for 1000 epochs, requiring approximately 20 hours of compute time. The networks for stress and ξ_m in (S4.4) are multilayer perceptrons (MLPs), both comprising four hidden layers with widths 256, 512, 512, and 256, using SELU activations and spectral normalization. In total, the model contains 1,060,107 trainable parameters. Pretraining of the RNO, by minimizing (S4.5), is performed jointly across the three models $k \in \{II, III, IV\}$ as follows. Recall that the encoders of the RNO are model independent while the decoders $\mathcal{F}_k$ and $\mathcal{G}_k$ are model dependent. The final two hidden layers of each of the MLPs are the decoders $\mathcal{F}_k$ and $\mathcal{G}_k$ and in each epoch the following 3 steps were performed:

- (i) Given material model $k = II$ we updated the parameters of the encoders $h_{\mathcal{F}}^{\text{RNO}}$ and $h_{\mathcal{G}}^{\text{RNO}}$ as well as the decoders $\mathcal{F}_{k=II}$ and $\mathcal{G}_{k=II}$.
- (ii) Next material model $k = III$ was selected and again the parameters of $h_{\mathcal{F}}^{\text{RNO}}$ and $h_{\mathcal{G}}^{\text{RNO}}$ were updated along with the parameters of decoders $\mathcal{F}_{k=III}$ and $\mathcal{G}_{k=III}$.
- (iii) Finally material model $k = IV$ was selected and again the parameters of $h_{\mathcal{F}}^{\text{RNO}}$ and $h_{\mathcal{G}}^{\text{RNO}}$ were updated along with the parameters of decoders $\mathcal{F}_{k=IV}$ and $\mathcal{G}_{k=IV}$.

Thus, in each epoch the parameters of $h_{\mathcal{F}}^{\text{RNO}}$ and $h_{\mathcal{G}}^{\text{RNO}}$ were updated three times and at the end of the optimisation we obtain 3 RNOs, one each for material models $k \in \{II, III, IV\}$. The

² The optimum choice of $d_\xi = 5$ internal variables is justified in Section S4.4.

encoders $h_{\mathcal{F}}^{\text{RNO}}$ and $h_{\mathcal{G}}^{\text{RNO}}$ are common between these 3 models while the decoders $\mathcal{F}_k$ and $\mathcal{G}_k$ are model specific. The evolution of the training (S4.5) with epochs is plotted in Fig. S19 for the $k = II$ material model. After 1000 epochs the loss reaches $\sim 1.4\%$. The evolution of the loss is very similar for all the three material models and hence for the sake of clarity only material model $k = II$ is shown in Fig. S19. These optimal pre-trained RNO models in Stage 1 were then selected for fine-tuning using experimental data in Stage 2.

S4.3 Finetuning with sparse experimental data (Stage 2)

In stage 2 we finetune the pretrained RNO using the curated experimental MT3.0 dataset (i.e. after correcting to ensure that the Drucker's stability criteria are satisfied; see Supplementary S2) of the notched Ti90 specimen (Fig. S3b) subjected to tension. Again, we first augment the experimental dataset and then using this augmented dataset to fine tune the RNO.

S4.3.1 Generation of experimental trajectories and data augmentation

We selected the 16 stress versus strain trajectories (Fig. S4) for finetuning the RNO. The plane stress trajectories directly available from the measurements are $\sigma_{11} - \varepsilon_{11}$ and $\sigma_{22} - \varepsilon_{22}$ after application of the Drucker stability correction (Supplementary S2.1) and $\sigma_{12} - \varepsilon_{12}$ where σ_{12} is inferred from the MLP (Supplementary S2). The plane stress assumption implies the stresses and strains $\varepsilon_{23} = \varepsilon_{13} = \sigma_{23} = \sigma_{13} = \sigma_{33} = 0$ and thus it only remains to specify ε_{33} to complete the fully 3D stress versus strain trajectories in the 16 locations. The elastic strain $\varepsilon_{33}^{\text{el}}$ follows from plane stress Hooke's law as

$$\varepsilon_{33}^{\text{el}} = -\frac{\nu}{E}(\sigma_{22} + \sigma_{11}), \quad (\text{S4.14})$$

and the in plane elastic strains are

$$\varepsilon_{11}^{\text{el}} = \frac{\sigma_{11}}{E} - \nu \frac{\sigma_{22}}{E}, \quad (\text{S4.15})$$

and

$$\varepsilon_{22}^{\text{el}} = \frac{\sigma_{22}}{E} - \nu \frac{\sigma_{11}}{E}. \quad (\text{S4.16})$$

Plastic incompressibility then requires that the plastic strain $\varepsilon_{33}^{\text{pl}}$ is given by

$$\varepsilon_{33}^{\text{pl}} = -[(\varepsilon_{22} - \varepsilon_{22}^{\text{el}}) + (\varepsilon_{11} - \varepsilon_{11}^{\text{el}})], \quad (\text{S4.17})$$

and ε_{33} then follows from (S4.14) and (S4.17) as

$$\varepsilon_{33} = \varepsilon_{33}^{\text{el}} + \varepsilon_{33}^{\text{pl}}. \quad (\text{S4.18})$$

We then have 16 full 3D stress versus strain trajectories from the notched tensile specimen and an additional uniaxial tensile response (Fig. S16). These trajectories were up/downsampled to 49 steps to match the loading step resolution at which the RNO was pretrained. These experimental trajectories occupy only a narrow region of the multi axial stress space, so we densified the dataset using the permutation augmentation described in Section S4.2 to generate a total of 102 trajectories.

S4.3.2 Finetuning protocol

The finetuning of the pretrained RNO with the augmented experimental dataset involved 2 steps: (i) selecting the RNO model to finetune and (ii) hyperparameters selection.

(i) *Model selection: proximity to the target*

Among the three pretrained material models $k \in \{II, III, IV\}$, we select the model whose predictions are closest to the experimental loading paths *before* any finetuning. For each material $\psi_k^\dagger$ we evaluate the error (S4.8) between the experimental trajectories $p = 1, \dots, P = 102$ with stresses $\sigma_{ij}^{*(p)}$ and the corresponding predictions $\hat{\sigma}_{ij}^{(m,k)}$ of model $\psi_k^\dagger$. Using this notation, we rewrite (S4.8) as

$$\mathcal{L}_S^{(k)} = \frac{1}{P} \sum_{p=1}^P \frac{\|\hat{\sigma}_{ij}^{(p,k)} - \sigma_{ij}^{*(p)}\|_F}{\|\sigma_{ij}^{*(p)}\|_F}, \quad (\text{S4.19})$$

where we have replaced $\hat{\sigma}_{ij}^{*(p)}$ in (S4.8) by $\hat{\sigma}_{ij}^{(p,k)}$ to emphasize that $\hat{\sigma}_{ij}^{(p,k)}$ is the output of the pretrained model $\psi_k^\dagger$ with no finetuning. The model $\psi_k^\dagger$ with the lowest $\mathcal{L}_S^{(k)}$ is chosen which in the case of Ti90 is the $k = II$ model with $\mathcal{L}_S^{(k)} \sim 10.1\%$. We select this model to finetune with the following justification. Finetuning is a local modification of the pretrained operator, and the smaller the initial residual, the smaller the parameter excursion required to fit the experimental data — and therefore the lower the risk of overwriting the inductive biases that the pretraining has already encoded.

(ii) *Hyperparameter selection and model selection*

The pretrained model $k = II$ has an error $\mathcal{L}_S^{(k)} \sim 10.1\%$ with respect to the 102 augmented experimental stress-strain trajectories. Given the relatively small experimental dataset it is critical to finetune the model with care to avoid overfitting which will result in the operator losing the inductive biases of the physics of plasticity that the pretraining has encoded. We selected the following hyperparameters over which to study the performance of the finetuned model:

- (i) *Learning rate*: We damped the pretraining learning rate r_{pre} in the finetuning step to $r_{\text{FT}} = \alpha r_{\text{pre}}$ where $0 \leq \alpha \leq 1$. A small α keeps the optimiser in the basin of the pretrained solution.
- (ii) *Layer freezing*: Finetuning only optimises the parameters of the decoders $\mathcal{F}_*^{\text{RNO}}$ and $\mathcal{G}_*^{\text{RNO}}$. However, again given the small dataset we do not optimize all the layers of this decoder but rather freeze f_{FT} and g_{FT} fully connected layers of $\mathcal{F}_*^{\text{RNO}}$ and $\mathcal{G}_*^{\text{RNO}}$, respectively. Freezing the shared early layers preserves some key features learned about the specific materials mode as only tails of $\mathcal{F}_*^{\text{RNO}}$ and $\mathcal{G}_*^{\text{RNO}}$ are updated by gradient descent.
- (iii) *Epochs*: The number of epochs E_{FT} is chosen to be large enough that the loss trajectory plateaus but not large enough to result in overfitting.

We performed a grid search over these parameters and obtained losses $\mathcal{L}_{\text{FT}}$ given by (S4.8) to lie in the range $3.75\% \leq \mathcal{L}_{\text{FT}} \leq 9.94\%$. We observed that the models with the lowest errors resulted in violations of the Drucker stability criterion under certain strain paths. Thus, trade-off between a low error $\mathcal{L}_{\text{FT}}$ and the model retaining the inductive physical biases is essential for a usable constitutive model. We chose a model with $\mathcal{L}_{\text{FT}} = 4.48\%$ obtained with finetuning parameters $\alpha = 0.1$, $f_{\text{FT}} = 0$, $g_{\text{FT}} = 1$ and $E_{\text{FT}} = 2000$.

S4.3.3 Predictions of the finetuned model

Selected predicted stress versus strain histories of the finetuned RNO compared to the measurements are included in Figs. 3c and 4c. In addition, in Fig. 3c we also show the RNO predictions of the yield surface on the π -plane. In the conventional Lou et al. [21] plasticity

model, the yield strength is a well-defined quantity as given by the yield surface Φ . However, the RNO structure is fundamentally different and is given by (S4.2), where stress versus strain trajectories are direct outcomes of the imposed loading history, and plastic strain or yield strain are not directly defined. To plot the yield surface, we follow a protocol similar to that used in experiments [28], viz. we load the material under a given strain path and measure the resulting stress versus strain curve. The yield strength is then defined as the stress at 0.2% from the initial linear elastic response. We numerically generated such trajectories using the finetuned model, and one such history (for all 6 stress components) is shown in Fig. S20a along with the defined 0.2% offset yield strength. Using 101 such trajectories we plot in Fig. S20b the yield surface as predicted by the finetuned RNO model on the π -plane. Observe that the predicted yield surface satisfies the isotropy encoded into the pretrained model (i.e. the yield surface is invariant to permutations in the principal stresses). We also confirmed that the RNO predicted yield surface is invariant to a superimposed pressure.

The advantage of the RNO over a conventional plasticity model is clear. It makes no a priori assumptions on the functional form of the yield surface, the flow rule or the hardening law, and can therefore captures complex plastic behaviour — including the non-monotonic stress responses and large plastic-strain increments beyond yield observed in the Ti90 measurements that conventional parameterised models (e.g. Lou et al. [21] or J2) struggle to reproduce. On the curated MT3.0 trajectories the RNO achieves an error $\mathcal{L}_{FT} = 4.48\%$ across the full measured stress–strain curves at every material point, compared to $\mathcal{R}_\sigma = 13\%$ for the best-fit Lou et al. [21] model calibrated on the same data; see Table S1 and note that $\mathcal{R}_\sigma$ and $\mathcal{L}_{FT}$ are analogously defined via (S3.11) and (S4.8), respectively. Moreover, the RNO validation errors using the compressive notched specimen and tensile complex specimen are also less than that of the best-fit Lou et al. [21] model (Table S1).

S4.4 Justification of the choices of our transfer learning approach

There are 2 key claims implicit in the RNO model described: (i) the number of internal variables $d_\xi = 5$ is sufficient and optimum and (ii) pretraining the model with multiple material models improves the performance of the RNO. We now justify these two claims.

S4.4.1 Optimum number of internal variables

We varied the number of internal variables d_ξ to investigate the ability of the RNO model to predict plastic responses. For this purpose, we split the augmented synthetic data comprising 3 materials models $k \in \{II, III, IV\}$ so that pretraining was conducted using models $k \in \{III, IV\}$ and $k = II$ used for finetuning. This 2-stage training strategy was performed for RNO models with $0 \leq d_\xi \leq 20$. The optimised finetuned loss $\mathcal{L}_{FT}$ is plotted in Fig. S21a as a function of d_ξ . The loss saturates for $d_\xi \geq 5$ confirming that $d_\xi = 5$ internal variables are sufficient to capture the path-dependence of plasticity of the family of yield surfaces considered here.

S4.4.2 Importance of pretraining

To demonstrate the importance of our transfer learning approach we now report an ablation study where we attempt to develop an RNO to capture the behaviour of a target synthetic dataset described by 20 stress versus strain trajectories. This target synthetic dataset was generated using a J2 plasticity model (i.e., equivalent to a Lou et al. [21] model with parameters $c = d = 0$) and the elastic and strain hardening properties of Ti90 (Table S3). With this choice of (c, d) the yield surface is distinct from material models $k \in \{II, III, IV\}$; see Fig. S21b.

We consider the following 2 cases:

- (i) Our preferred transfer learning approach outlined above where we pretrain the RNO with the 3 material models $k \in \{II, III, IV\}$. Then using the J2 model the losses for the three pretrained models were $\mathcal{L}_S^{(II)} = 36\%$, $\mathcal{L}_S^{(III)} = 37\%$ and $\mathcal{L}_S^{(IV)} = 32\%$. Based on this we selected model $k = IV$ for finetuning with 20 target J2 stress versus strain curves. A test error $\mathcal{L}_{FT} = 8.3\%$ was obtained.
- (ii) We randomly initialised the RNO parameters and performed no pretraining and only finetuned with the target 20 J2 stress versus strain trajectories. In this case we obtained an error $\mathcal{L}_{FT} = 32\%$.

Thus, we observe a $\sim 4 \times$ reduction relative to the from-scratch baseline with our multi-material pretraining.

S4.5: Structural finite element (FE) calculations using the finetuned RNO

The finetuned RNO described in Section 4.3 is integrated into a quasi-static implicit finite-element framework MOOSE (Multiphysics Object-Oriented Simulation Environment) [29] as a custom user material with the neural operator evaluated through LibTorch, the PyTorch C++ API [30, 31]. The network parameters are fixed during the finite-element solution, while the recurrent latent variables are stored as material state variables at each quadrature point of the FE mesh. At each material update, the RNO takes the current strain input together with the previously converged internal state and returns the updated stress and internal state. The tangent supplied to the nonlinear solver is the local algorithmic Jacobian of this implemented RNO update with respect to the current strain input. We compute this Jacobian directly using PyTorch/LibTorch automatic differentiation, rather than finite differences. In this differentiation, the previous converged state is held fixed, but the derivative still accounts for the dependence of both the updated internal state and the stress on the current strain. Thus, the finite-element solver receives a tangent that is consistent with the discrete RNO material update evaluated in MOOSE.

The finite element calculation of the notched and complex specimens shown in Figs. 3b, 4d and S9 were performed using 8-noded 3D brick elements. The specimens were meshed with 2 elements through the specimen thickness and a graded mesh to capture large gradients near the features such as notches and holes. The remarkable accuracy of the RNO predictions for two complicated structural settings confirms that the RNO, trained using measured data, is sufficiently robust to perform structural calculations and therefore for use in engineering design.

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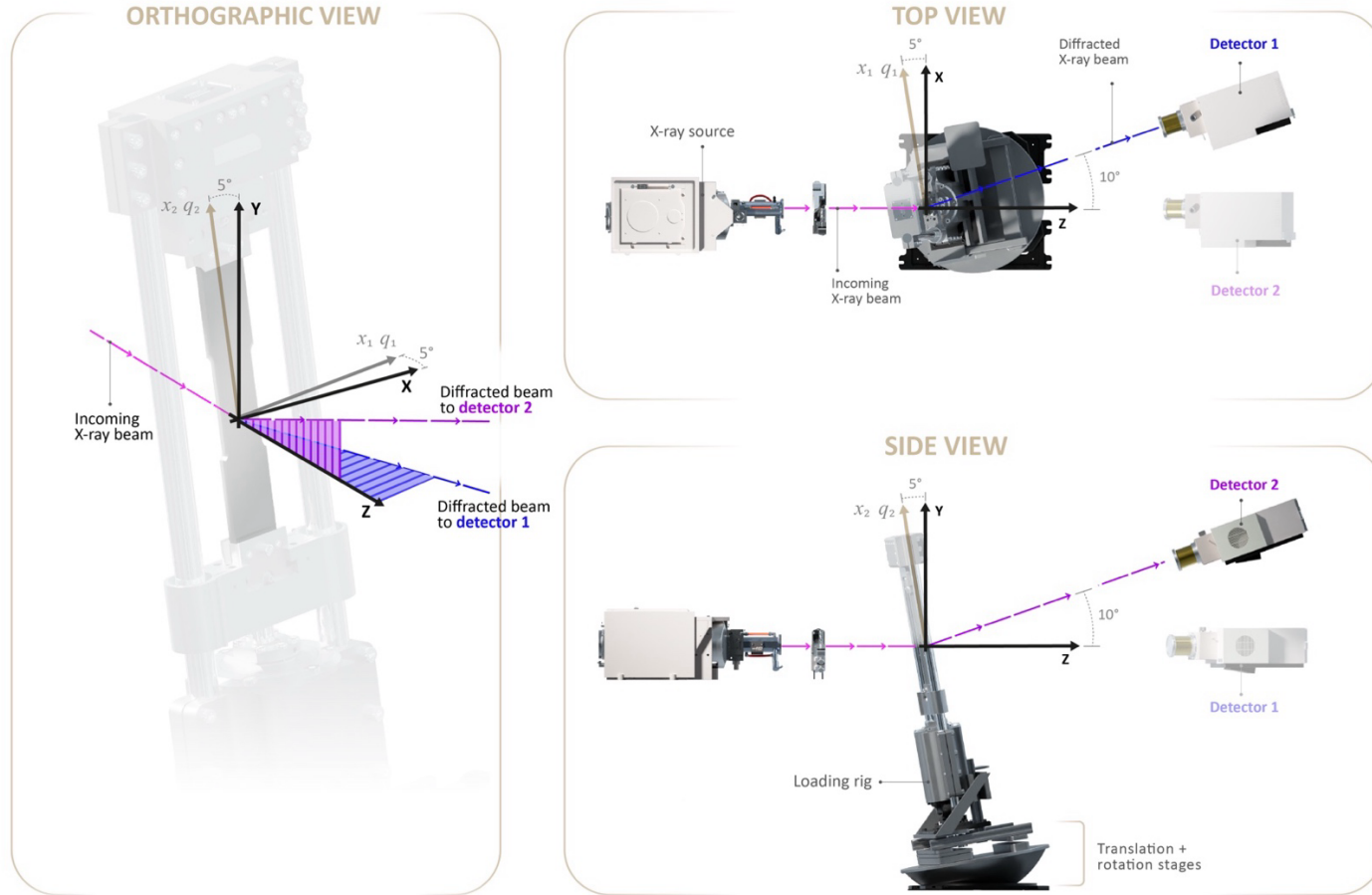


Figure S1: A view of the load frame from Fig. 1b (not to scale) to illustrate its orientation for elastic strain measurements. The (X, Y, Z) co-ordinate system fixed with respect to the incoming X-ray beam is shown to clarify the rotation $\theta = 5^\circ$ of the load frame about the X and Y –directions. The specimen with the (x_1, x_2, x_3) co-ordinate system fixed on the specimen with x_2 aligned with the loading direction is also shown. The q –vectors q_1 and q_2 along which the lattice strains are measured via diffraction (and set by the direction of the incoming X-ray beam and the detector locations) are also indicated along with the diffracted beams at $2\theta = 10^\circ$ with respect to the incoming X-beam (Z –direction). The top and side views clarify the orientations of the loading system for the q_1 and q_2 vectors, respectively.

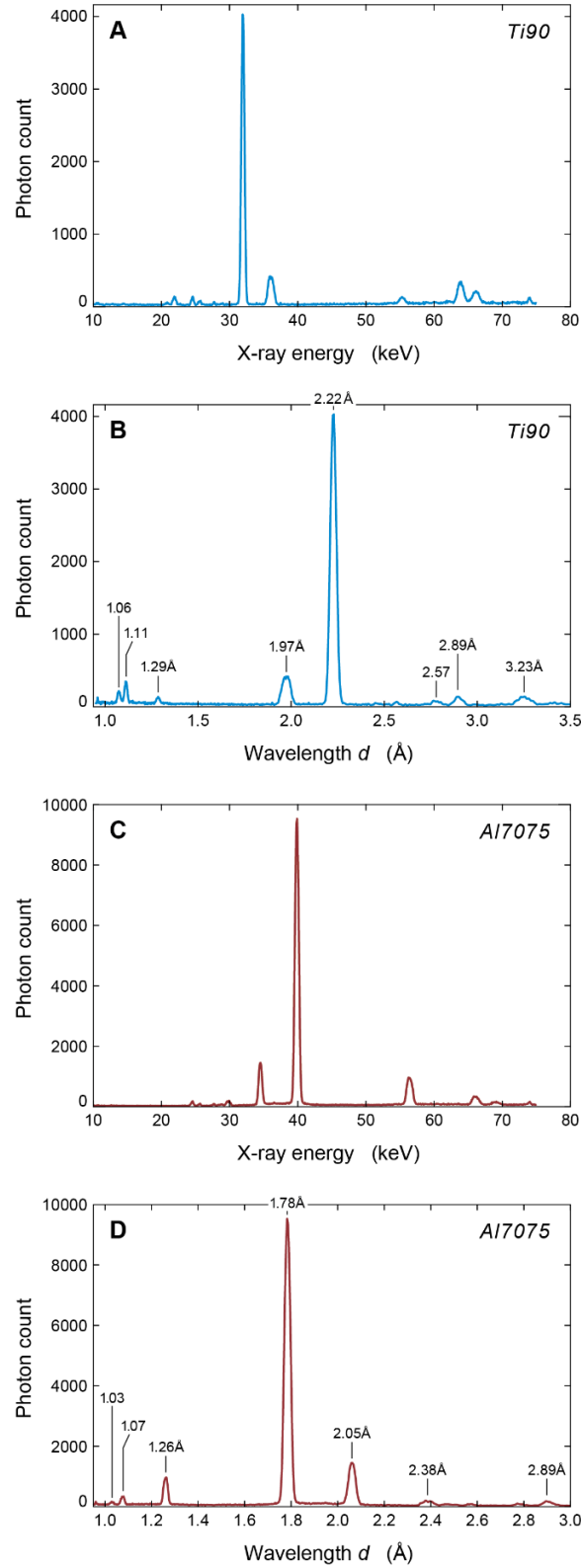


Figure S2: The measured EDXRD spectra for the (a,b) 0.5 mm thick Ti90 and (c,d) 2.5 mm thick Al7075 with a 18000 s photon collection time. These spectra were measured on the detector that gives the d -spacings in the q_2 vector direction (although the isotropy of the materials implied that these spectra are identical for the 2 detectors). In each case we show the spectra as a function of X-ray energy (i.e. raw output from the detectors) and converted to d -spacing using Bragg's law with the glancing angle taken to be $\theta = 5^\circ$.

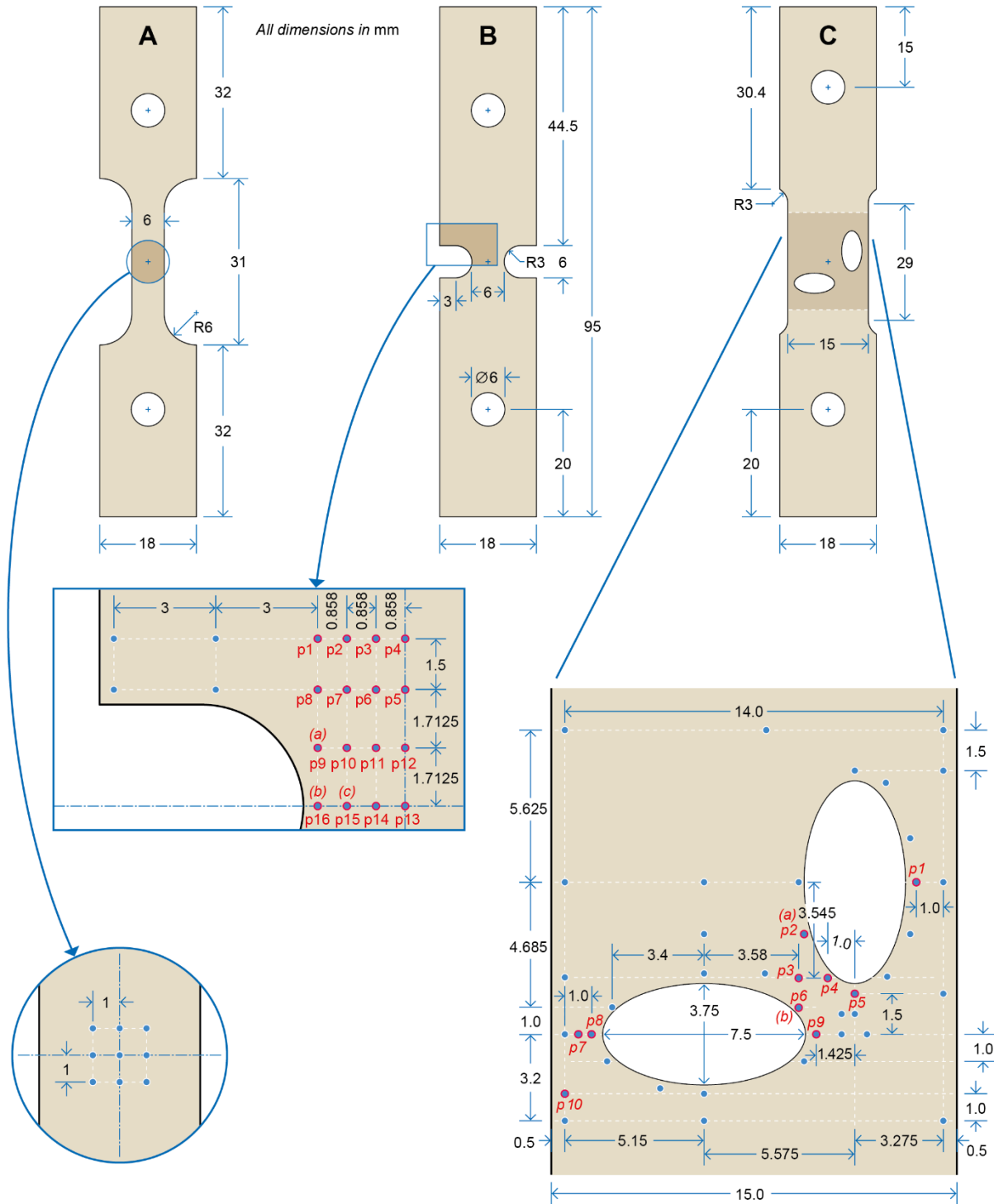


Figure S3: Sketches of the 3 specimen geometries with leading dimensions marked. In all three cases we also indicated the locations where the EDXRD measurements were conducted. Key dimensions are labelled and the figures drawn to-scale to enable positioning of all marked material locations. (a) The uniaxial specimen, (b) notched specimen and (c) specimen with a complex geometry. In (b) and (c) we have labelled the material point locations where the measured stress versus strain curves are plotted in Figs. S4, S6 and S8 as well as the material points at which curves are plotted in Figs. 3 and 4. These labelled points plus the other points marked are the full set of material locations where EDXRD measurements were conducted to enable plotting of the stress contours shown in Figs. 3b and 4d. All dimensions are in mm.

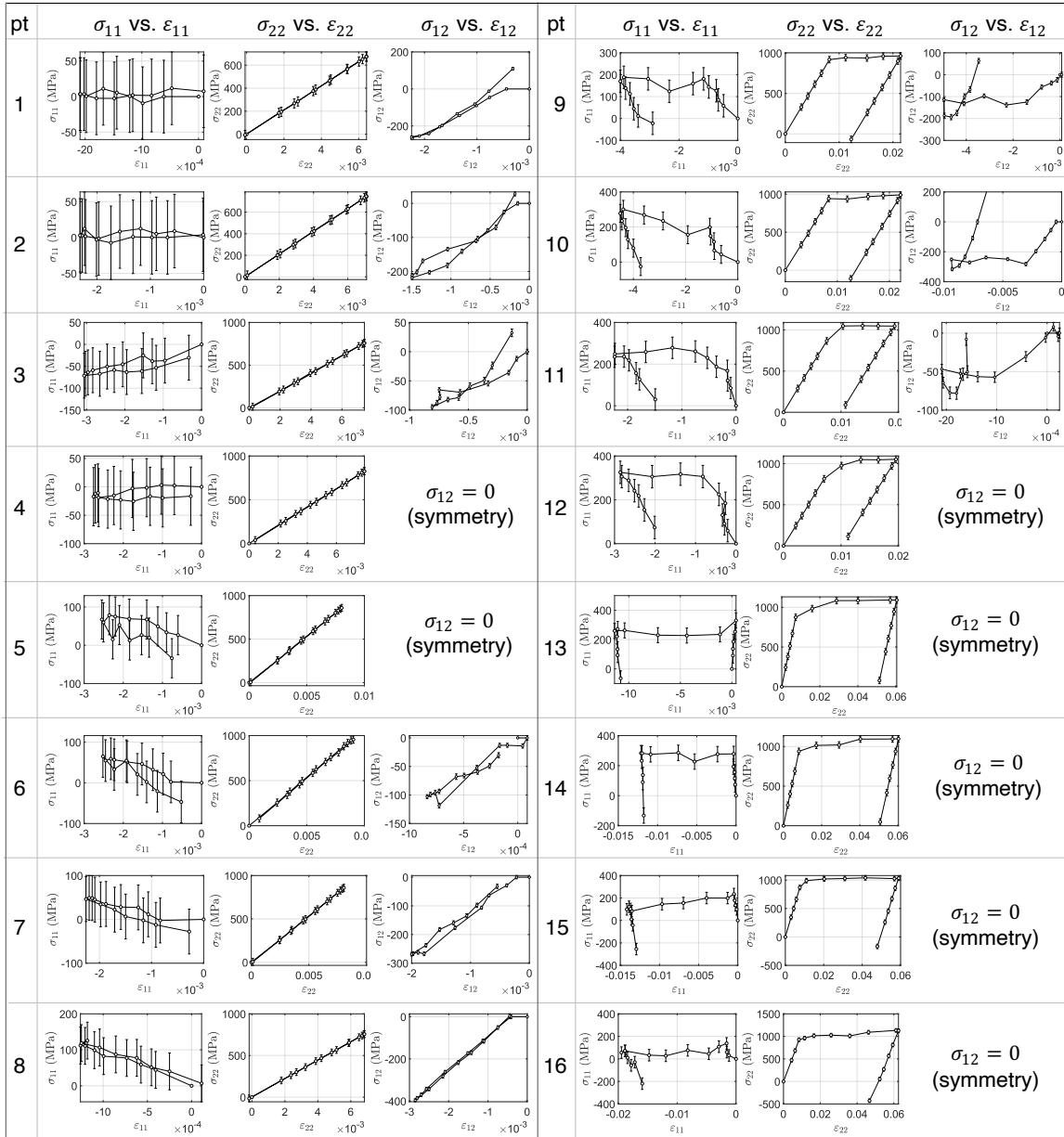


Figure S4: The stress versus strain curves measured during the tensile test on the notched Ti90 specimen (Fig. 3b). Curves are shown at 16 material locations as marked and labelled in Fig. 3b and in each case we include estimates of uncertainties as error bars. For locations on the plane of symmetry the shear stress $\sigma_{12} \approx 0$ and hence the shear stress versus strain curves are excluded.

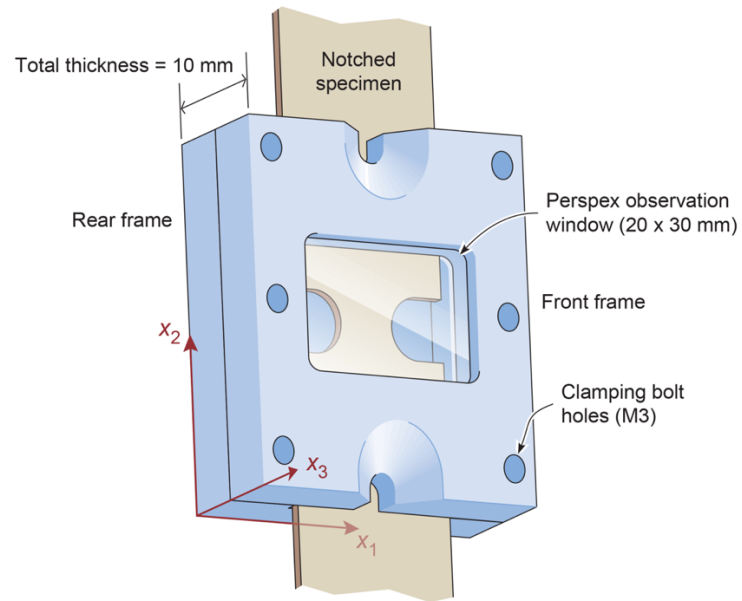


Figure S5: Sketch of the anti-buckling guide assembly for conducting compression tests on the thin plane stress specimens. The sketch shows this assembly with the notched specimen.

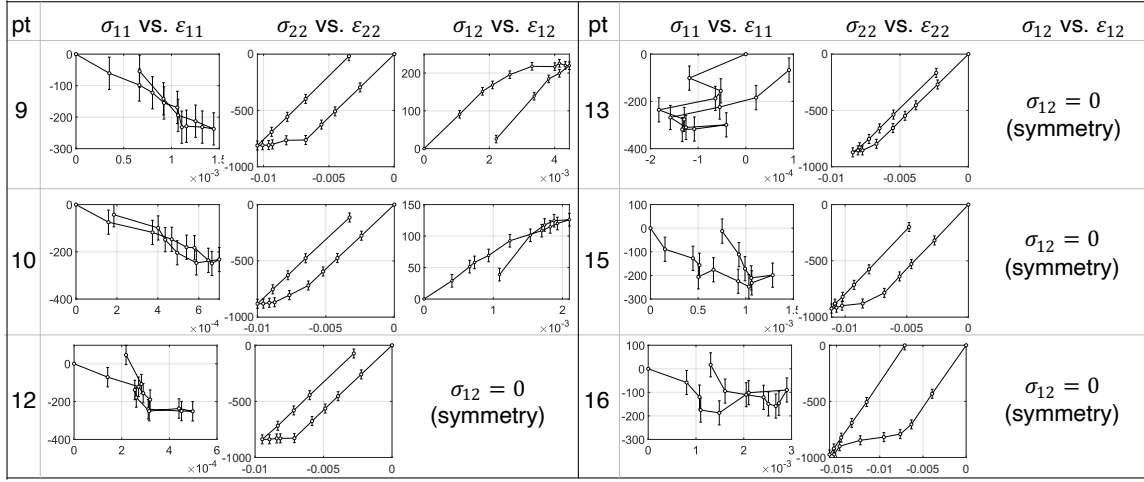


Figure S6: The stress versus strain curves measured during the compressive test on the notched Ti90 specimen (Fig. 3b) conducted with anti-buckling guides. Curves are shown at 6 material locations as marked and labelled in Fig. 3b and in each case we include estimates of uncertainties as error bars. For locations on the plane of symmetry the shear stress $\sigma_{12} \approx 0$ and hence the shear stress versus strain curves are excluded.

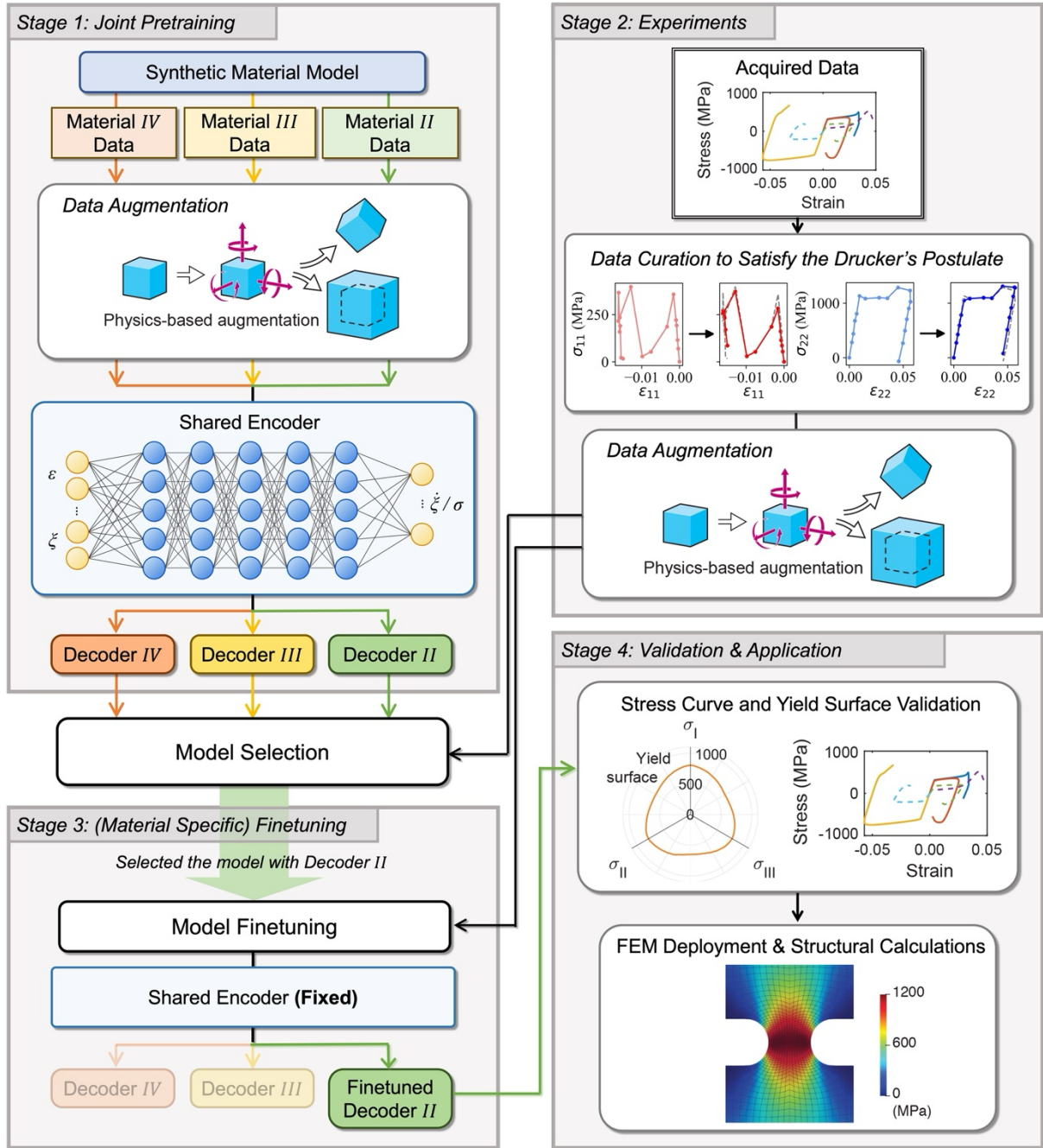


Figure S7: Flow chart to show the workflow of the transfer learning protocol to develop RNO plasticity models for metal plasticity from the MT3.0 measurement datasets.

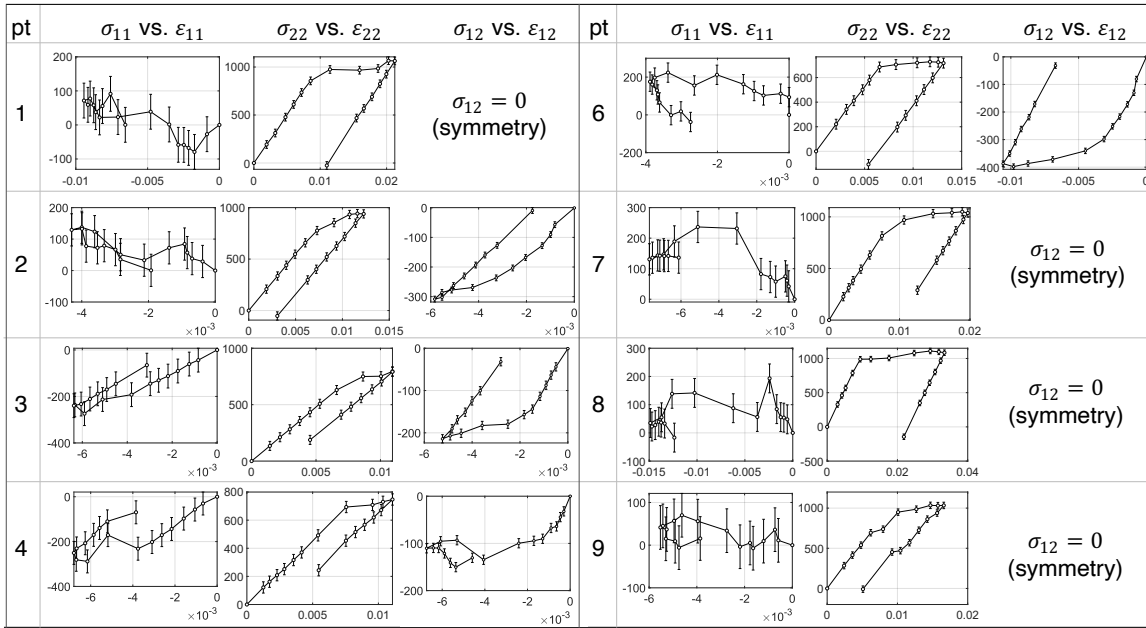


Figure S8: The stress versus strain curves measured during the tensile test on the complex Ti90 specimen (Fig. 4b). Curves are shown at 8 material locations as marked and labelled in Fig. S3c where significant plasticity was observed to occur. In each case we include estimates of uncertainties as error bars.

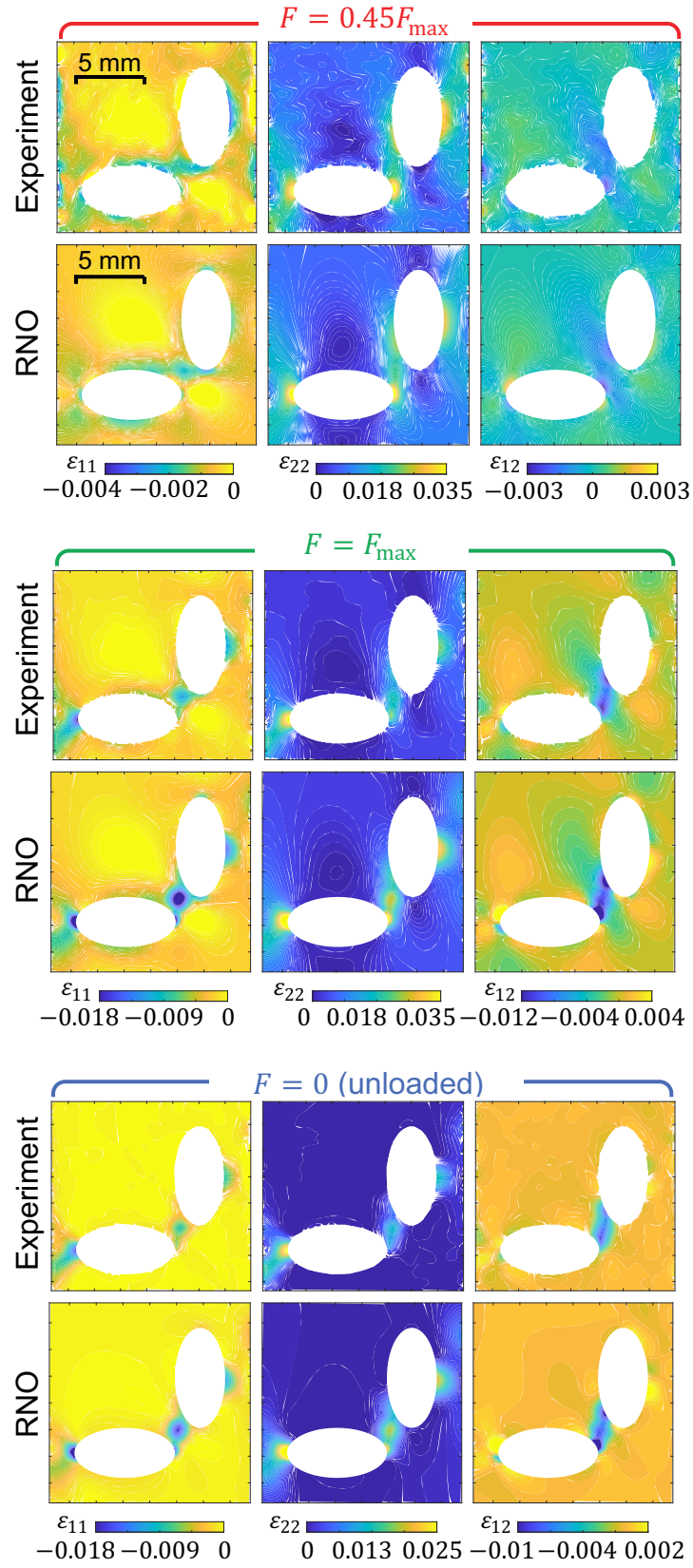


Figure S9: Comparison between the measurements and RNO predictions of strain fields in complex specimen at the 3 loading stages marked in Fig. 4b. These strain field correspond to the stress fields shown in Fig. 4d.



Figure S10: Photograph of the MT3.0 facility that has democratized synchrotron technology and brought combined EDXRD and 3D DIC measurement capabilities to everyday laboratory experiments.

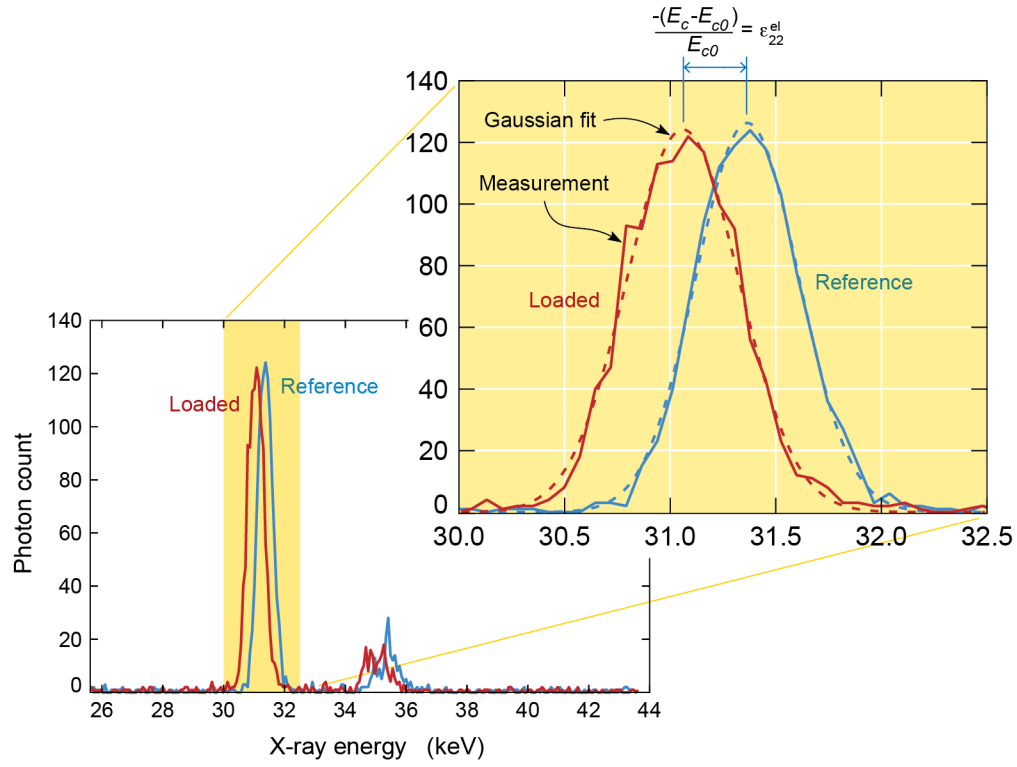


Figure S11: A representative EDXRD spectrum for a Ti90 specimen in its loaded and reference state with a 200 s photon collection time. The spectrum is from the detector associated with the q –vector q_2 . Only the 2 largest energy peaks are included in the spectrum. The Gaussian fits (Section S1.3) are included and the shift of the mean peak positions from the unloaded to loaded state for the largest peak indicated.

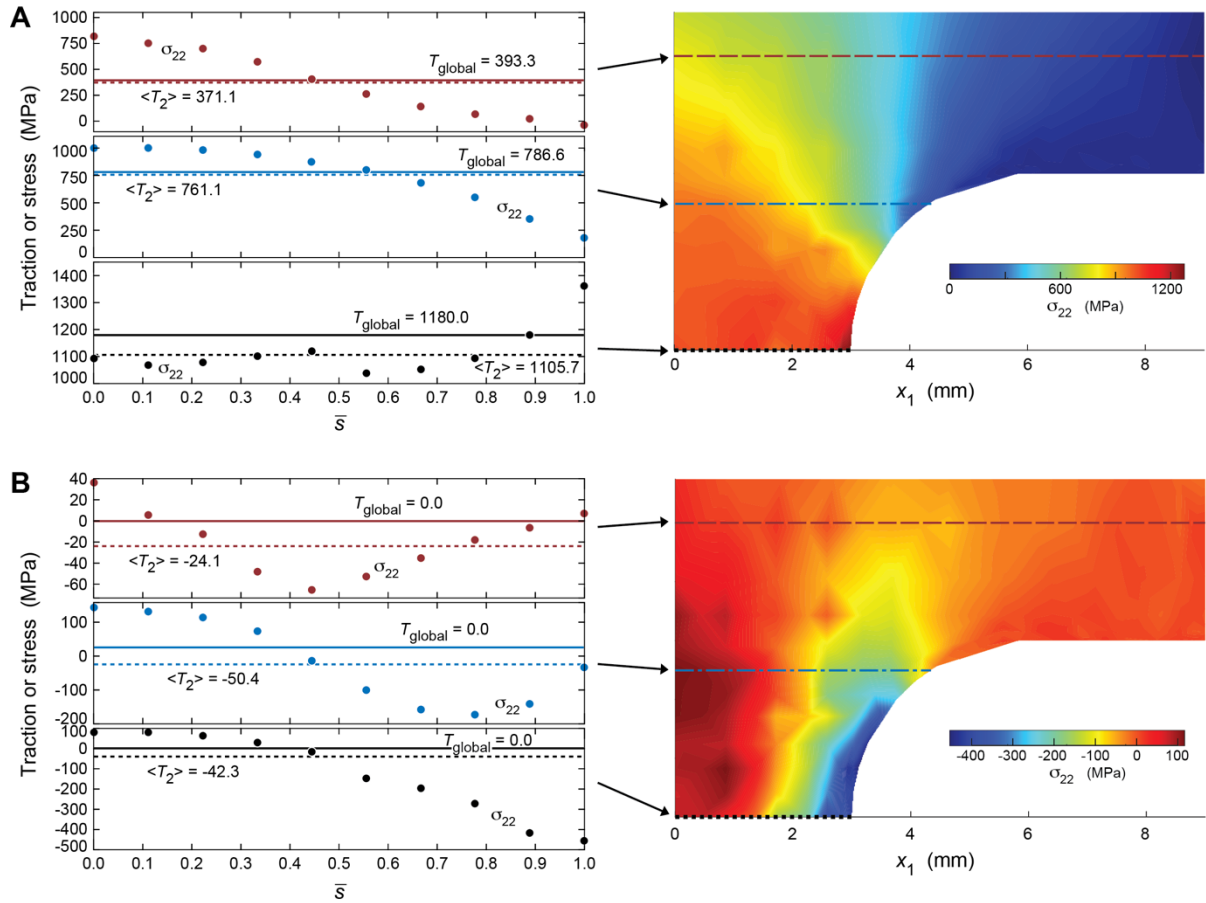


Figure S12: Verification of the EDXRD measurements for the notched specimen. The distributions of stress σ_{22} in the specimen (a) at peak load and (b) after unloading to zero load, i.e. residual state. The locations of the 3 lines along which tractions T_2 are then inferred are marked. The distributions of the EDXRD stresses σ_{22} (data points) as well as the mean EDXRD traction $\langle T_2 \rangle$ and mean traction $\bar{T}_{\text{global}}$ (in MPa) from the measured global load are included for the 3 lines in both (a) and (b).

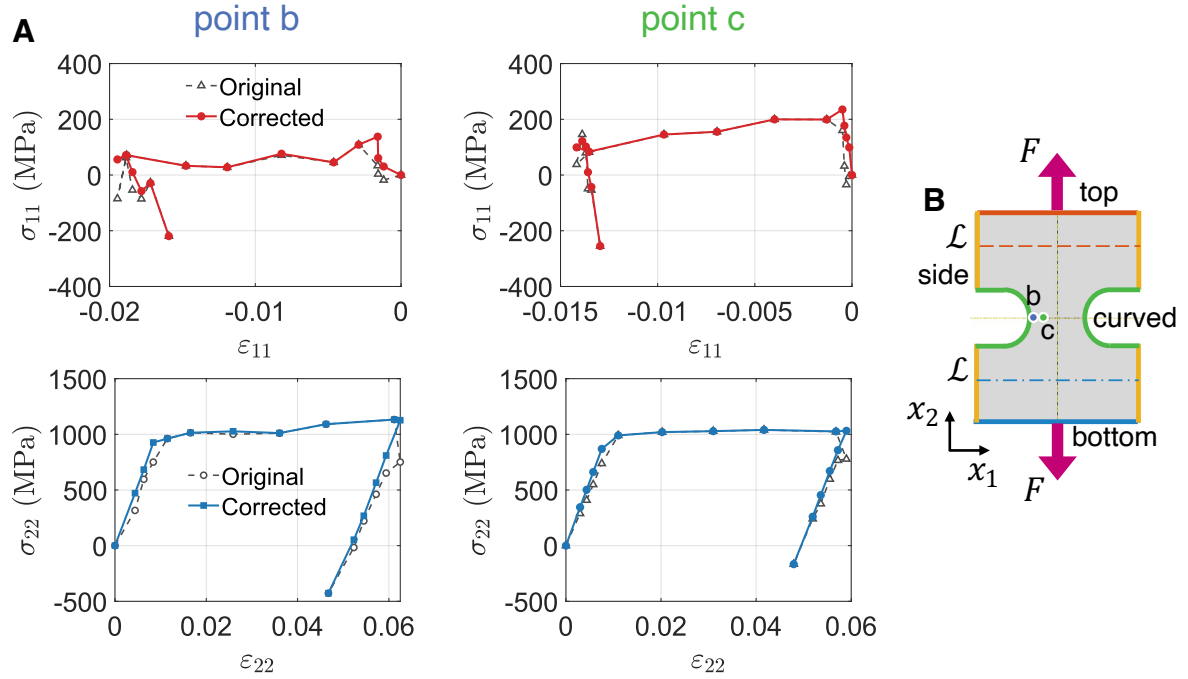


Figure S13: (a) Illustration of the measured $\sigma_{11} - \epsilon_{11}$ and $\sigma_{22} - \epsilon_{22}$ curves at material point “b” and “c” in the notched specimen subject to tensile loading (Fig. 3a and Fig. 3c) prior to and after correction to ensure that the Drucker’s stability criterion is not violated. (b) A sketch of the notched specimen the “side” and “curved” surfaces and “top” & “bottom” surfaces labelled along with the co-ordinate system (x_1, x_2, x_3) . The specimen is loaded by a force F along x_2 with equilibrium then demanding that the total force along the x_2 -direction is also F for any line $\mathcal{L}$ along x_1 . The approximate locations of material points “b” and “c” are also indicated.

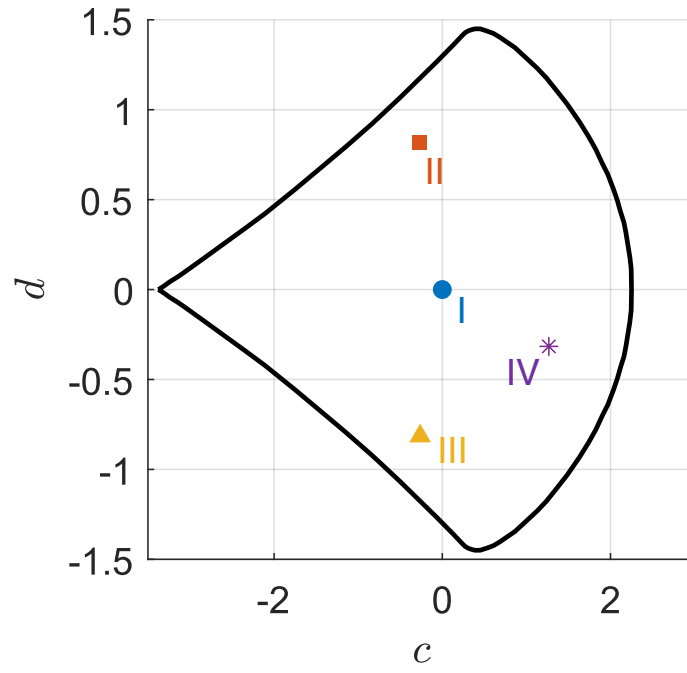


Figure S14: The domain of parameters (c, d) for the yield function Φ given by (S3.2) to remain convex.

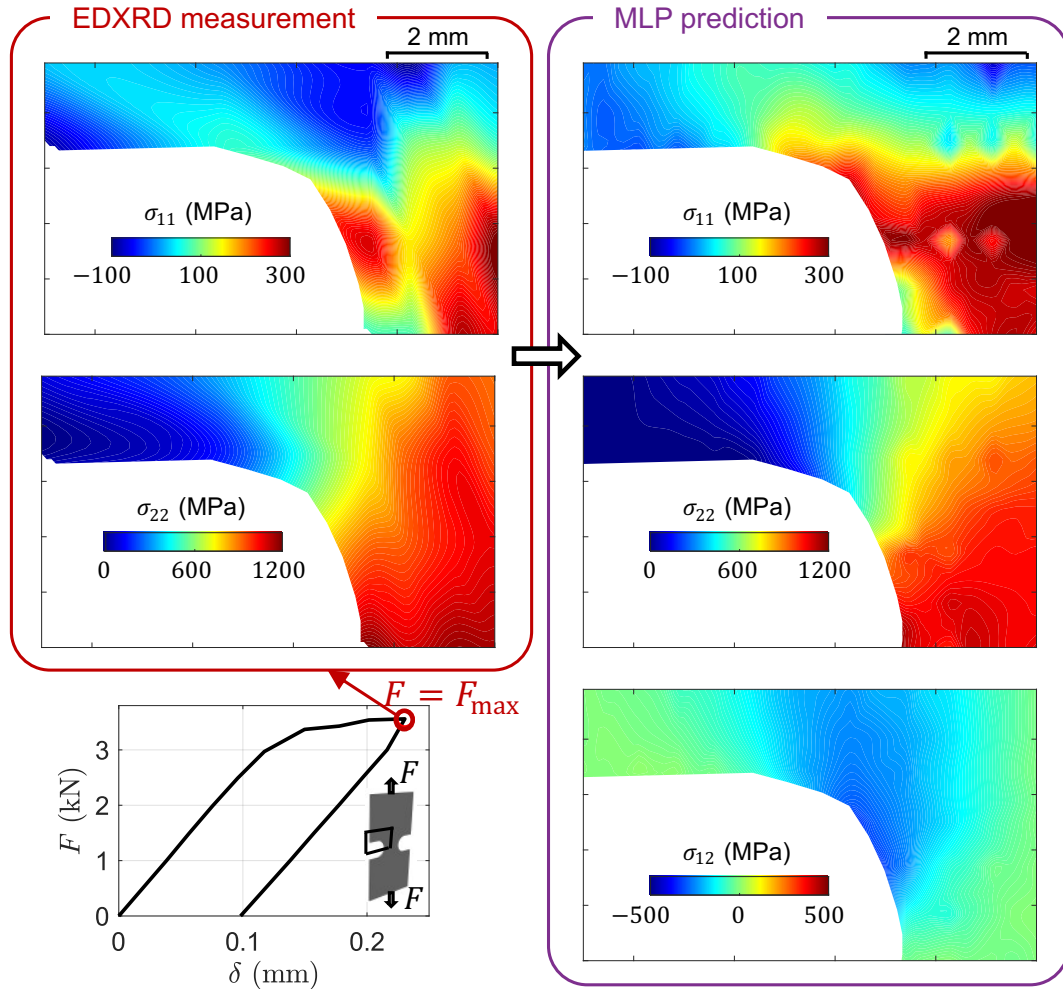


Figure S15: Distributions of the stresses (σ_{11} , σ_{22} , σ_{12}) as predicted by the MLP in the notched specimen at peak tensile load. These are the same distributions shown in Fig. 3b. The EDXRD measurement is shown as a contour plot using linear interpolation of the stresses between the measurement locations.

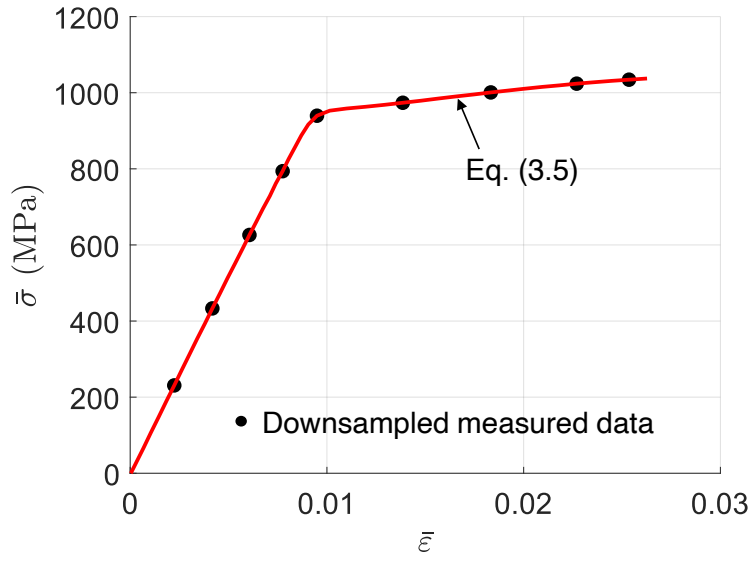


Figure S16: The measured uniaxial tensile stress $\bar{\sigma}$ versus strain $\bar{\epsilon}$ curve of the 0° Ti90 specimen. A best fit response (S3.5) is included using the parameters listed in Table S3.

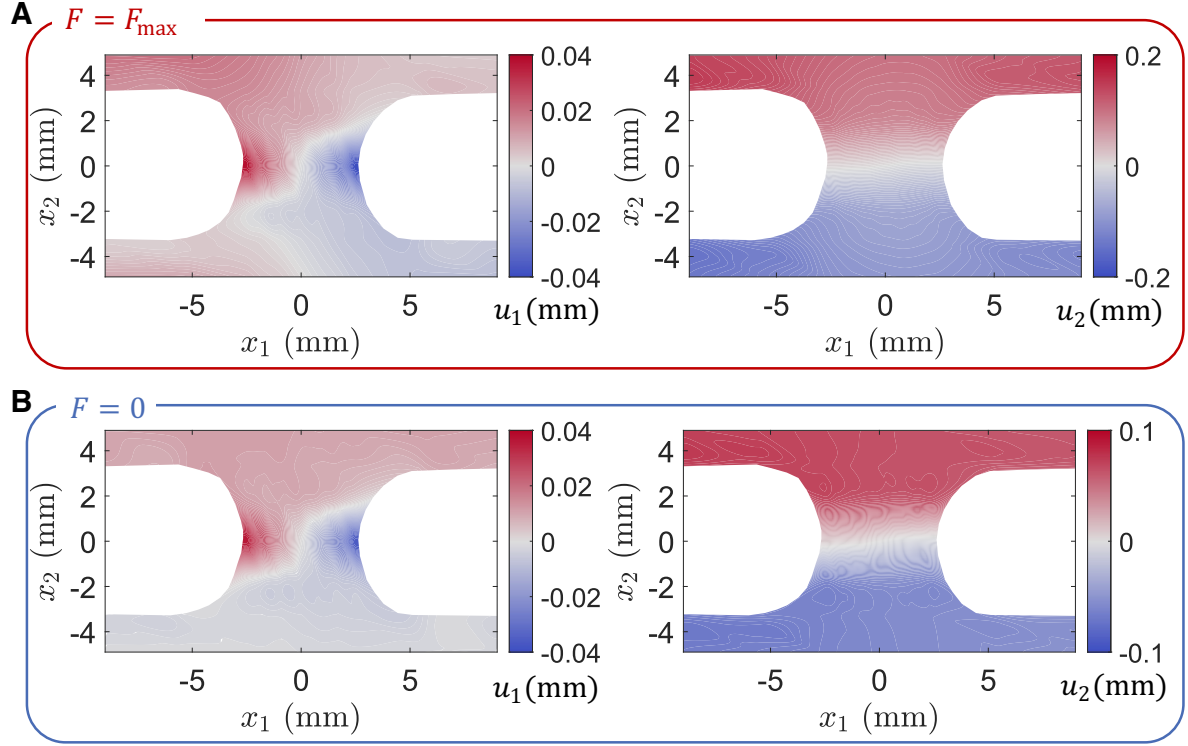


Figure S17: An example of the displacement data used in the MT2.0 protocol for the notched tensile Ti90 specimen. The 3D DIC measurements of the displacements (u_1, u_2) within the gauge section of the specimen at (a) maximum load $F_{\max}$ and (b) after unloading to $F = 0$.

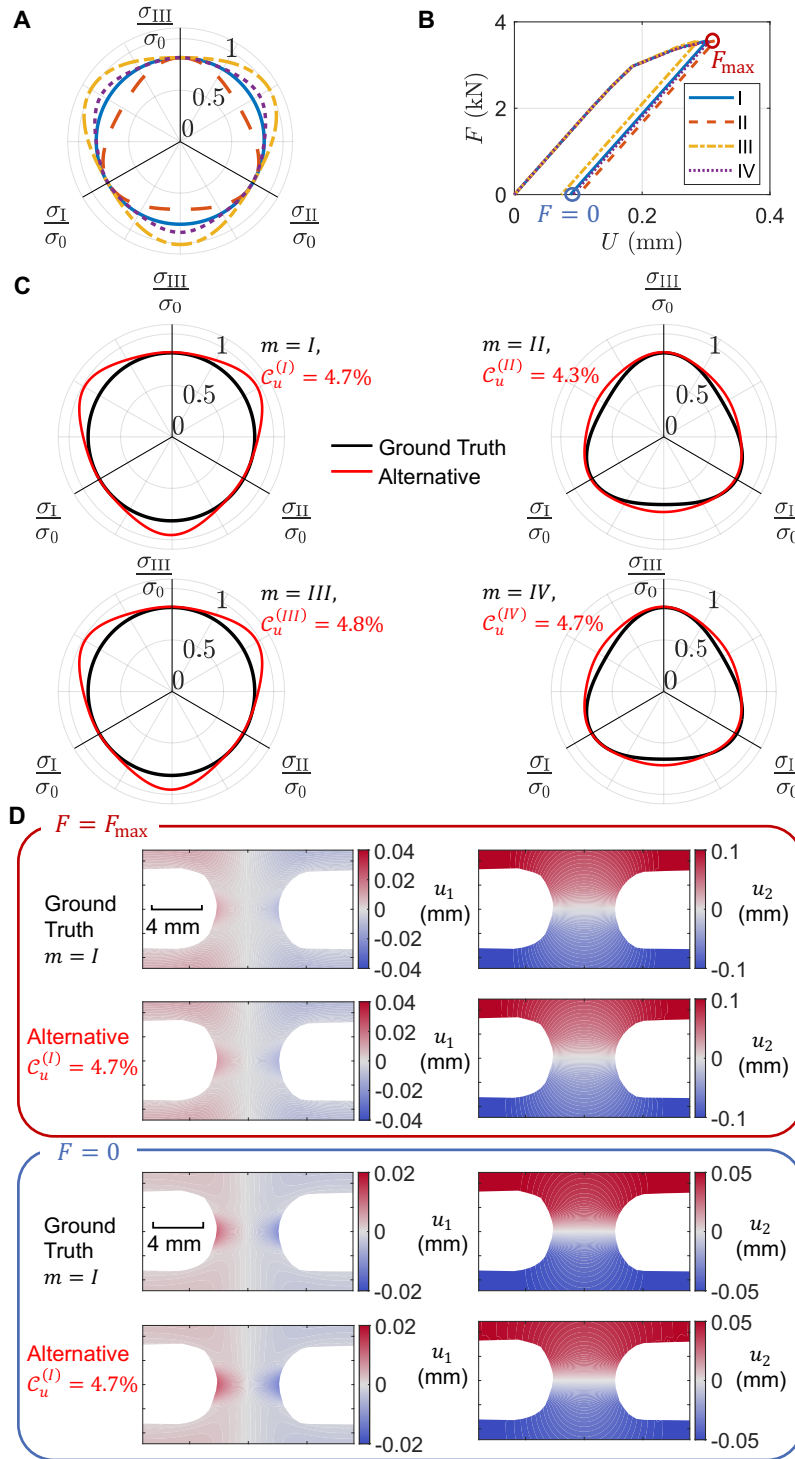


Figure S18: (a) The four yield surfaces ($m = I, \dots, IV$) with different types of asymmetries used to illustrate the ill-posedness of the MT2.0 protocol. The parameters (c, d) for these 4 yield surfaces are listed in Table S5. (b) The FE predictions of the applied tensile load F versus displacement U response of the notched specimen made from materials following these 4 different yield surfaces. (c) The true (black) and alternative choice (red) of the yield surface for each choice $m = I, \dots, IV$. In each case the alternate yield surface has an error $\mathcal{C}_u^{(m)} \leq 5\%$. (d) A comparison between the predicted distributions of the displacement u_i using the J2 yield surface ($m = I$) and an alternative surface with $(c = 1.0, d = -0.6)$ that gives an error $\mathcal{C}_u^{(I)} = 4.7\%$.

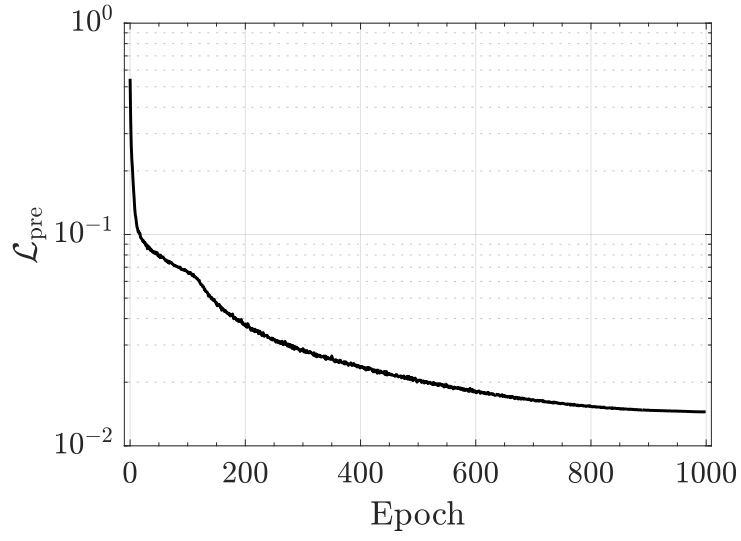


Figure S19: Evolution of the pretraining loss (S4.5) with epochs for pretraining of the $k = II$ material model. These curves are very similar for the other material models $k = III, IV$ and hence omitted here for the sake of clarity.

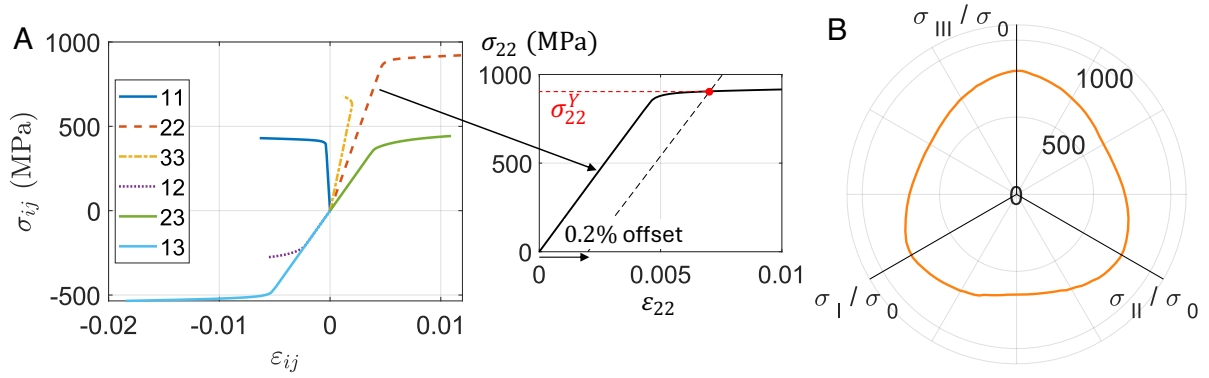


Figure S20: (a) Prediction of the stress versus strain history for the finetuned RNO model for a selected monotonic strain path. Curves are shown for all 6 stress and strain components, and the inset shows a sketch to demonstrate how the 0.2% offset yield strength for the σ_{22} component (labelled σ_{22}^Y) is defined using the $\sigma_{22} - \varepsilon_{22}$ trajectory. (b) The predicted RNO yield surface based on the 0.2% offset yield strength definition. The yield surface is shown on the π -plane.

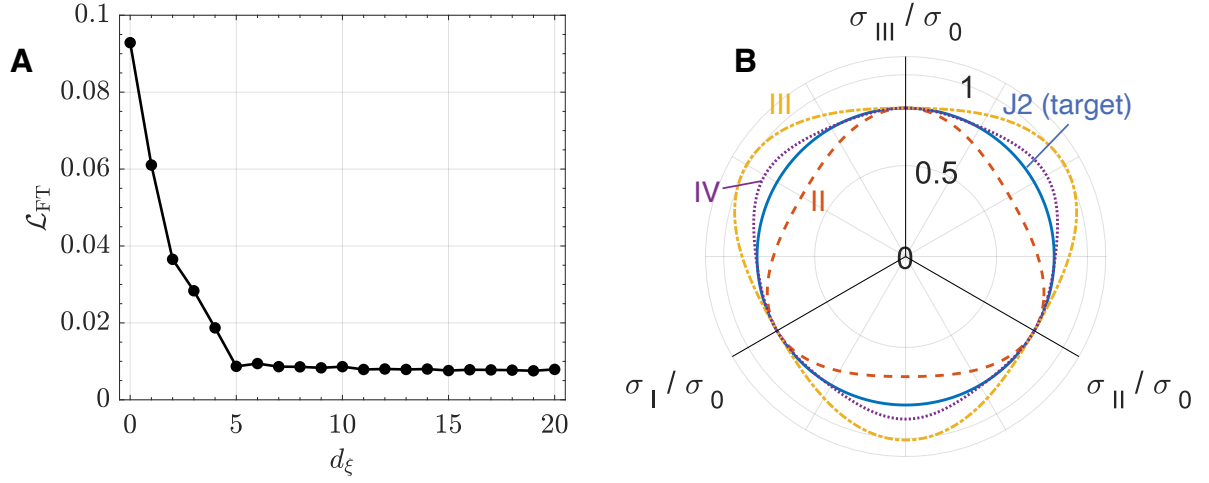


Figure S21: (a) The predicted optimised finetuned loss $\mathcal{L}_{FT}$ for a synthetic dataset as a function of the number of internal variables d_ξ . The RNO was pretrained on models $k \in \{III, IV\}$ and finetuned on model $k = II$ (Table S5). (b) A comparison between the yield surfaces for Lou et al. [21] models $k \in \{II, III, IV\}$ and J2 model with Lou et al. [21] parameters $(c, d) = (0, 0)$ used in the study to demonstrate the importance of multi-material pretraining.

Supplementary Tables

Protocol	Notched tension	Notched compression	Complex tension
$\mathcal{R}_\sigma$ for Lou et al. [21]	13.0%	14.6%	14.7%
$\mathcal{L}_{FT}$ for RNO	4.5%	9.5%	14.6%

Table S1: The $\mathcal{R}_\sigma$ (for Lou et al. [21]) and $\mathcal{L}_{FT}$ (for RNO) models calibrated using the notched tensile specimen data. We show three cases: (i) the notched tensile specimen for which the models were calibrated and therefore $\mathcal{R}_\sigma$ and $\mathcal{L}_{FT}$ optimised and the corresponding validation errors of the models in predicting the material responses in the (ii) compressive notched specimen and (iii) the tensile complex specimen. The data is obtained using the MT3.0 protocol. Note that $\mathcal{R}_\sigma$ (S3.11) and $\mathcal{L}_{FT}$ (S4.8) are analogously defined but given different notation to clearly differentiate between them.

	R_0	R_{45}	R_{90}	R_n
$\bar{\epsilon} \sim 0.015$	1.028	0.971	1.052	1.005
$\bar{\epsilon} \sim 0.025$	1.017	0.980	1.020	0.999

Table S2: The Lankford coefficients R_0 , R_{45} , R_{90} and R_n of the Ti90 sheets employed in this study. These coefficients are listed for two values of the imposed tensile strain $\bar{\epsilon}$.

Material parameter	Symbol	Value
Young's modulus	E	105 GPa
Poisson's ratio	ν	0.33
Initial yield strength (S3.5)	σ_0	935 MPa
Strain hardening rate (S3.5)	h_σ	1.8 GPa
Strain hardening exponent (S3.5)	n	0.7

Table S3: The elastic and strain hardening parameters for Ti90.

Protocol	c	d	$\mathcal{C}$	$\mathcal{R}_\sigma$
MT2.0	2.13	-0.42	11%	15%
MT3.0	-0.53	0.65	12%	13%

Table S4: The Lou et al. [21] parameters (c, d) predicted by the MT2.0 and MT3.0 protocols along with the associated optimal errors $\mathcal{C}$ and $\mathcal{R}_\sigma$ for the notched tensile specimen.

Material choice label m	c	d
I (J2- plasticity)	0	0
II	-0.2669	0.8161
III	-0.2633	-0.8147
IV	1.2670	-0.3160

Table S5: The 4 choices of the Lou et al. [21] parameters (c, d) used to demonstrate the ill-posedness of the MT2.0 protocol. These 4 choices are labelled $m = \text{I, II, III, IV}$.

Supplementary Movie

Movie S1: Illustrative movie of the MT3.0 facility and its key components, including the high-flux X-ray source, polychromatic X-ray beam collimation system, in situ mechanical loading stage, three-dimensional digital image correlation (3D-DIC) system, and high-efficiency X-ray detectors. The facility democratizes energy-dispersive X-ray diffraction (EDXRD) by enabling laboratory-scale experiments that were previously possible only at large-scale synchrotron facilities, thereby bringing advanced multiaxial materials characterization capabilities into standard research laboratories.