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Observing deformation in situ

Marc Legros, Frédéric Mompiou and Daniel Caillard discuss the different aspects that influence the reproducibility and reliability of characterizations performed using in situ mechanical tests in transmission electron microscopes.

Mechanical in situ transmission electron microscope (TEM) experiments consist of deforming a piece of material in a TEM while capturing the nanoscale mechanisms responsible for its plastic behaviour, correlated with microstructures such as dislocations, twins and phases. The goal is to observe how matter deforms while measuring the contribution of each mechanism, to understand how the mechanisms combine and thus ultimately explain macroscopic mechanical properties. With the development of better imaging tools, micromechanical holders and dedicated numerical methods, this goal finally seems within reach. In practice, however, the gap that separates the macroscopic scale from the micro-/nanoscopic scale and the complexity of microstructures make the reproducibility of such tests questionable, with numerous caveats.

One of the first concerns is the interaction of the powerful electron beam (typically 100–300 keV) with the sample, potentially changing its structure. Besides the moderate heating generated by this interaction (at the origin of the first observation of a moving dislocation[1]), and the potential irradiation damage to light metals that constituted an early concern[2], recent studies have reported a drastic alteration to the mechanical properties of amorphous silica[3] and more moderate changes in aluminium or gold[4]. The physical reasons for such recently reported variations are still unclear, but technical and scientific advances including those described below should provide adequate tools with which to unravel them.

With the advent of focused-ion-beam milling, TEM sample preparation has gained universality and precision but has also generated worries regarding structural modifications, because the gallium ions both implant in the TEM sample and cause irradiation damage. Such damage can create or interact with dislocations, modifying the mechanical properties of the crystal being studied[5]. Such damage also hinders the observation of 'intrinsic' defects, precluding a clean characterization of a given microstructure[6]—an ubiquitous but rarely mentioned problem. Although techniques exist to remove such preparation artefacts, the electropolishing of bulk metals is usually considered the safest method of avoiding structural modifications.

Even with minimum modification, free surfaces may influence a given microstructure through image forces that tend to attract the dislocations to the surface, favouring their escape from the crystal, their dissociation or the rate of kink nucleation along their lines. The contribution of image forces depends on several factors such as surface properties (oxidation and so on), studied phenomena and sample thickness. Although it is generally weaker than the dislocation confinement effect, there is no general criterion to quantify the influence of surface forces, which means that it should be discussed and evaluated for each experiment.

The first in situ technique to appear consisted of a miniaturized tensile test using an infinite screw-type holder in which no load is measured (a conventional holder). These holders operate on millimetre-sized TEM samples, usually electropolished, offering a transparent area of several square micrometres. Moreover, their stiffness is several orders of magnitude higher than that of the TEM sample, so that burst-type events such as dislocation avalanches and Lüders slip bands can only be attributed to intrinsic factors of the material. Such events are frequently observed in nanomechanical holders such as the in situ nanoindenters that appeared in the 2000s. These tools include piezo-electric actuators and miniaturized load cells that allow stress measurements, sometimes at the expense of the rigidity of the whole. Strain bursts like those visible in the stress–strain curve of Fig. 1a (ref. 7) may be characteristic of small-scale plasticity, but may also be a result of the holder compliance or the slow feedback of its electronic control unit.

These nanoindenter-type holders are often claimed to be the first ones able to generate stress–strain curves in a TEM, but early developments in high-voltage electron microscopy[8] did so using conventional holders in the early 1970s. Such established technologies also have the advantage of allowing temperature control (from liquid helium to very high temperatures[9]) over a range currently inaccessible using piezo-based nanoindenters. Another complication inherent to compression is the very fast degradation of imaging conditions during in situ tests (partly visible in Fig. 1b). One reason is that TEM imaging conditions are very sensitive to the small crystal rotations that inevitably occur during compression, owing to buckling, for instance. Dark-field imaging, as employed in Fig. 1b, is a way to overcome this, but the transformation of compression to tension using 'push to pull' lithography-patterned silicon devices[9] could be a more versatile option. Indeed, straining under tension helps to maintain imaging conditions by stabilizing the orientation of the microsample after the yield.

In all cases, technical improvements are needed to improve in situ TEM tests to levels that meet the standards of the American Society for Testing and Materials. The use of micro-electromechanical systems may be one way to reach this goal, but again, this will only be possible in very specific samples (those made by focused-ion-beam milling, nanostructures and thin films). Potential charge effects on capacitance readouts (displacement and load) should also be controlled[10].

The disadvantage of not having load cells on conventional tensile straining holders may become an advantage because the strain increments are perfectly controlled. Larger accessible volumes and steady imaging conditions also offer better statistics on dislocation behaviour, and imaging quality allows measurements of the behaviour of single dislocations, using machine learning recipes[11]. A common substitute for external load cell measurement is the use of dislocation

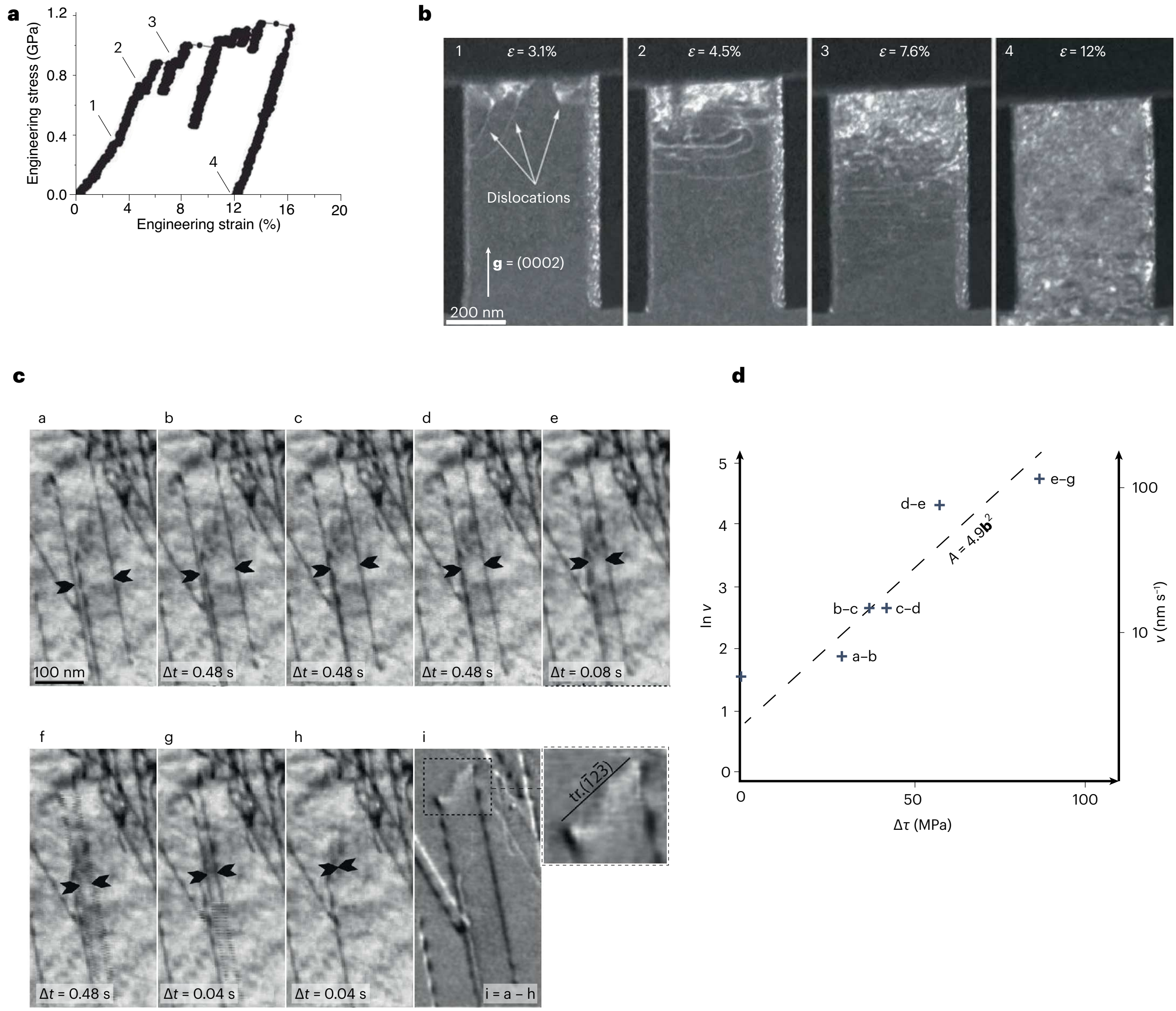

Fig. 1 | Dynamical sequences captured during in situ TEM strain or compression experiments and associated mechanical properties. **a**, Stress–strain curve obtained from the in situ compression of a magnesium micropillar along its *c* axis using a nanoindenter sample holder[7]. **b**, Dark-field TEM snapshots 1–4 obtained with diffraction vector **g** corresponding to arrows 1–4 in **a**, showing dislocations emitted from the top of the pillar at various stages of compression strain (ε = 3.1–12%). **c**, In situ TEM sequence a–h, where Δt is the time interval between two successive images, obtained using a conventional straining holder on electropolished high-purity iron, showing a screw dislocation dipole (black arrows) annihilation at 113 K. Image i is the difference between images a and h. The enlargement of the selected area in image i shows the direction of the average slip trace (tr.) at the foil surface. **d**, The activation area A of the Peierls mechanism that governs the motion of dislocation in body-centred cubic crystals, directly inferred from such a test. The slope of the natural log of the velocity $\ln v$ versus the elastic interaction $\Delta\tau$ gives a direct value of the activation area for single dislocations[14] (the stresses and velocities marked a–h correspond to a–h in **c**); **b** is the Burgers vector of the dislocation. Panels **a** and **b** adapted with permission from ref. 7, AAAS; panels **c** and **d** reproduced with permission from ref. 14, Elsevier.

curvature as a local stress probe[12], an approach that matches macroscopic yield stress measurements in iron[13] and several other metals and alloys as a function of temperature, considering the small increase of stress due to the confinement effect in the thin foil.

Figure 1c,d also exemplifies how a dislocation dipole annihilation can be turned into a perfect single-dislocation experiment[14]. There, both the elastic interaction stress and the velocity increase as the dislocations come close to each other, allowing a direct and fully local assessment of the activation volume linked to the Peierls motion of these screw dislocations in the body-centred cubic structure of iron[14]. Such access to intrinsic dislocation properties is unique to in situ TEM experiments, although very seldom exploited.

These experiments could also provide a direct benchmark for atomistic simulations, either molecular dynamics or discrete dislocation dynamics[15].

Finally, in situ TEM strain publications often report only single experiments, which is far from the scientific requirement to repeat an observation to fully validate its occurrence. In samples or nanostructures made using focused-ion-beam milling, the limited volume accessible to dislocation, or their interaction with interfaces and obstacles, shrinks the available information even more. An obvious reason for this lack of experimental repetition is that the success rate of such experiments is often very low. In any case, any extraordinary behaviour should require more scrutiny from the whole community before being accepted as valid and before extrapolations to a possible macroscopic property should be drawn from it.

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Competing interests

The authors declare no competing interests.