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Local, atomic-level elastic strain measurements of metallic glass thin films by electron diffraction

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ABSTRACT

A novel technique is used to measure the atomic-level elastic strain tensor of amorphous materials by tracking geometric changes of the first diffuse ring of selected area electron diffraction patterns (SAD). An automatic procedure, which includes locating the centre and fitting an ellipse to the diffuse ring with sub-pixel precision is developed for extracting the 2-dimensional strain tensor from the SAD patterns. Using this technique, atomic-level principal strains from micrometre-sized regions of freestanding amorphous $Ti_{0.45}Al_{0.55}$ thin films were measured during in-situ TEM tensile deformation. The thin films were deformed using MEMS based testing stages that allow simultaneous measurement of the macroscopic stress and strain. The calculated atomic-level principal strains show a linear dependence on the applied stress, and good correspondence with the measured macroscopic strains. The calculated Poisson's ratio of 0.23 is reasonable for brittle metallic glasses. The technique yields a strain accuracy of about 1×10^{-4} and shows the potential to obtain localized strain profiles/maps of amorphous thin film samples.

1. Introduction

The measurement of elastic strain is important for the analyses of physical properties of materials, e.g. the electronic properties of semiconductors. The miniaturization of electronic components and the interest in nanostructures necessitates strain characterization on a local scale. Therefore, in recent years several new techniques have been developed to measure lattice strains of crystalline materials at nanometre scale resolution using transmission electron microscopy (TEM) methods [1,2]. The techniques used to obtain quantitative information are based on either (i) TEM imaging like high resolution TEM [3] and dark-field electron holography [4] or (ii) TEM diffraction like nano-beam electron diffraction [5] and convergent beam electron diffraction [6].

In the case of amorphous materials, elastic strain and large scale strain distribution have been measured only by the use of X-ray diffraction [7–10] even though metallic glasses have also been studied intensively by TEM methods due to their unique mechanical properties [11–13]. The strain determination using X-ray diffraction is based on measuring the deviation of the broad diffraction rings from circular symmetry. This diffraction-based method enables the determination of the atomic-level elastic strain of amorphous materials whereas the macroscopic strain contains anelastic contributions as well [14].

In this paper we show that TEM selected area electron diffraction (SAD) can also be applied to analyze atomic-level elastic strains of metallic glass thin films. Using this method, we obtain the elastic strain tensor from micrometre-sized regions of freestanding, amorphous TiAl film samples during in-situ TEM tensile deformation. In addition, the elastic properties of the sample are also calculated by taking the applied stress into account. The determination of elastic strain requires an accurate analysis of the SAD patterns, which is implemented as a plug-in (SAD-strain [15]) written for the GATAN™ Digital-Micrograph platform. By acquiring diffraction patterns at different sample locations, we show that strain profiles or strain maps can be obtained on a micrometre scale using this method.

2. Atomic-level strain analysis of amorphous structure by electron diffraction

The diffraction pattern of alloys with an amorphous structure typically consists of a few diffuse rings (cf. Fig. 1a). If the amorphous material is isotropic the elastic scattering intensity $I(\boldsymbol{q})$ is only a function of the magnitude of the scattering vector $\boldsymbol{q}$ and not

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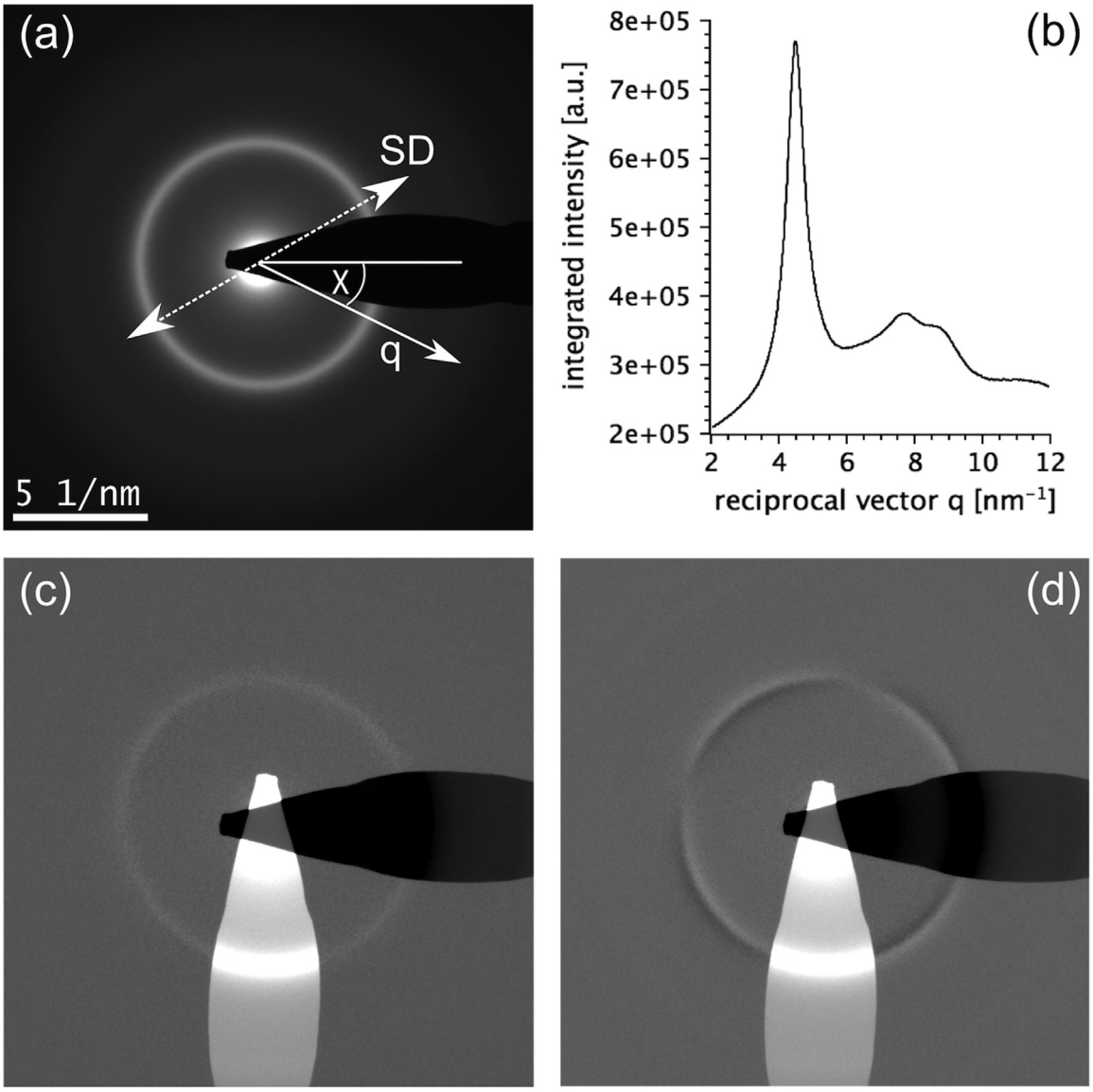

Fig. 1. Selected area electron diffraction (SAD) pattern and the average image of SAD patterns with their rotated and inverted counterparts. (a) SAD pattern of a TiAl thin film showing the first intense amorphous ring and the corresponding reciprocal lattice vector $\boldsymbol{q}$ at an angle χ. The straining direction (SD) is indicated in the image by the double arrow. (b) Intensity profile $I(q)$ obtained by integration of the intensity along rings. (c) SAD pattern overlay at zero stress reveals a circular symmetric diffuse amorphous ring. (d) Due to the ellipticity of the SAD pattern of the strained sample, a clear deviation from circular symmetry can be observed in the image overlay.

of the direction. This rotational symmetry of the intensity distribution allows the integration along rings (over the azimuthal angle χ) to obtain $I(q)$ (cf. Fig. 1b) as well as the structure function $S(q)$. The latter is calculated from $I(q)$ by taking into account the atomic scattering factors based on the composition of the material [9]. The circular symmetry of the SAD ring pattern (in the unstrained condition) is illustrated in Fig. 1c.

If the amorphous structure becomes anisotropic due to internal or external stresses, the diffraction pattern deviates from circular symmetry. This deviation from symmetry forms the basis for the atomic-level strain analysis of X-ray data, as suggested by Poulsen et al. [7]. During uniaxial tensile loading, the atoms will tend to move apart in the loading direction while in the transverse direction the atoms will move closer due to the Poisson's effect. These changes in real space distances lead to a shift of the intensity maxima in the diffraction pattern. The position q_1 of the first maximum is shifted to lower q-values in the tensile direction and to higher ones in the transverse direction. Hence, the overlay of a distorted SAD ring pattern with its rotated and inverted counterpart yields an image with a two-fold rotation axis only (cf. Fig. 1d). The relative change of the position of $q_1(\sigma,\chi)$ at a given stress σ with respect to the unloaded position $q_1(0,\chi)$ can be used to calculate the atomic-level strain $\epsilon(\sigma,\chi)$ from reciprocal space analysis as a function of the azimuthal angle χ as reported in [7,8,16]:

$$\epsilon(\sigma,\chi) = \frac{q_1(0,\chi) - q_1(\sigma,\chi)}{q_1(\sigma,\chi)} \tag{1}$$

In order to calculate small deviations from circular symmetry that represent strain, the positions of intensity maxima and the corresponding q_1-values have to be measured very accurately. Therefore, the centre of the diffraction pattern as well as the peak positions were measured with sub-pixel accuracy by data fitting procedures as explained below. These procedures are implemented as GATAN™ Digital Micrograph plug-ins, using the Armadillo library to easily handle matrices [17]. Additional features are implemented by calling script functions provided by the PASAD tools [18]. The algorithm performs the following steps of