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# In-situ observation of crack propagation through the nucleation of nanoscale voids in ultra-thin, freestanding Ag films

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ABSTRACT

A tensile technique was developed and coupled with in-situ transmission electron microscopy observations to directly characterize the crack propagation mechanism in sputter-deposited, ultra-thin, freestanding nanocrystalline Ag thin films with a thickness of 60 nm. The developed technique directly revealed the fracture mechanism; the thin film with nanoscale grains exhibits ductile fracture behavior, and the crack propagates through void nucleation, growth, and coalescence ahead of the crack tip. A model for the energy release rate during the propagation of nanovoids was established to quantitatively characterize the equilibrium length of the voids. Based on experimental measurements and theoretical calculations, the effects of stress distribution and energy transformation on the nucleation position, equilibrium length, and growth rate of the nanovoids are discussed.

## 1. Introduction

Compared to their coarse-grained counterparts, nanocrystalline (NC) thin films exhibit enhanced properties in terms of strength, hardness, and wear resistance [1–5]. These attributes have generated considerable interest in the use of these metallic systems with nanoscale grains for a wide variety of structural and functional applications. For example, thin films of face centered cubic (FCC) structured metals have been widely used in a variety of technologies, including flexible electronics [6–8], microelectromechanical systems (MEMS) [9], and deformable coatings. However, in most cases, NC thin films have poor ductility [10], which limits their practical utility. Therefore, it is crucial to understand the fundamentals of the fracture propagation mechanisms that are responsible for the brittle or ductile behavior of NC materials.

Over the last century, a number of milestone developments have been achieved in fracture mechanics at the macroscopic scale [11,12]. However, most of the characteristics that describe the behaviors of bulk materials frequently fail in describing the material response of thin layers [13]. The thin film considered in this study was NC, and the ultra-fine nanograins in this film have a considerable influence on its mechanical properties due to complex dislocation and grain boundary (GB) interactions [14]. Similar to the softening of NC metals with very small grain sizes [15], the effective strain hardening capacity also varies with the grain size [16,17]. Therefore, crack propagation in NC thin films is a complex physical process that spans several length scales, from atomistic to continuum scales, and NC thin films may differ from their coarse-grained counterparts with respect to their deformation behaviors.

The mechanism of crack propagation in thin films is of paramount importance for many thin-film applications. This topic has attracted considerable interest in engineering and the sciences, and a substantial amount of research has been conducted, including molecular dynamics simulations [18] and experimental studies [19], to uncover the mechanisms of fracture processes in thin films. Many potential mechanisms have been proposed, including conventional lattice dislocation slip [20], vacancy diffusion [21], and the emission of prismatic and shear dislocation loops [22].

However, previous studies regarding crack propagation were primarily based on molecular dynamic (MD) simulations or *ex-situ* studies. Many investigators have studied the properties of thin films on substrates [23] and have struggled with separating the properties of the film and substrate. Direct in-situ experimental observations and studies of crack tip propagation in freestanding films without substrates are limited due to difficulties in the fabrication and handling of NC freestanding thin films without damage and due to difficulties in performing in-situ observations at sufficiently high magnifications to follow local changes in the microstructure during deformation [24].

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Compared with other methods, the in-situ transmission electron microscopy (TEM) test, in which the specimens to be investigated are typically freestanding films, can provide more relevant information because this test directly observes crack propagation [25,26]. In addition, the freestanding tensile test is capable of directly observing the fracture mechanism of thin films without interfering effects from the substrate [27–30]. At present, the experiments on freestanding thin-films taken by in-situ TEM [31–37] are mainly focus on the deformation rather than fracture mechanisms.

To the best of our knowledge, the crack propagation behaviors in ultra-thin Ag films with thicknesses of less than 100 nm have rarely been observed using the freestanding tensile test. Therefore, the purpose of this study is to directly observe the crack propagation behavior at nano-scale resolution using an in-situ TEM study on crack propagation in freestanding nanoscale Ag thin films with an ultra-thin thickness of 60 nm and to suggest a theoretical model that quantitatively describes the nucleation and growth of nanovoids (NVs) in deformed NC Ag thin films.

In-situ experiments conducted in a TEM allowed the nanostructural behavior to be observed, which was then related to the dominant deformation mechanisms. It will be shown that although the nanoscale grain size had some impact on the deformation mechanism, the stress distribution is similar to that in the bulk material. The stress plays an important role in crack propagation, and the propagation of cracks proceeds through the nucleation, growth, and coalescence of voids. The quantitative description of the nucleation and growth of NVs may be helpful for interpreting the crack propagation mechanism. The details of this study are presented in the following sections.

## 2. Materials and experimental procedure

### 2.1. Nanocrystalline freestanding thin-film fabrication

To avoid the effect of surface oxidation, a NC Ag film was selected for the in-situ straining experiments. NC Ag thin films with a thickness of approximately 60 nm were sputter-deposited onto a $SiO_2/Si$ wafer using DC magnetron sputtering from a high-purity target ( $>$ 99.99%). The deposition was conducted using a substrate temperature of less than 40 °C and a base pressure of less than $5.0\times10^{-6}$ Torr. The Ag thin films were etched to obtain a uniform rectangular shape of 30 μm in length and 5 μm in width. The deposition rate was approximately 1.15 nm/s. The Ag films were transferred onto a carbon support film containing copper grid for the TEM analysis. The as-deposited material was examined in an electron microscope to confirm that the grain boundaries did not contain any amorphous layers, secondary phase particles, or nanoscale voids.

### 2.2. Microstructure of the as-deposited Ag film

Microstructural investigations were conducted using TEM techniques as shown in Fig. 1. The specimens were found to be dense with no voids or pores in either the grain interior or boundaries. The grain boundaries did not exhibit evidence of secondary-phase particles or films. The selected area diffraction pattern, which is shown as an inset in Fig. 1a, exhibits the expected ring pattern for such ultra-fine-grained materials, and analysis of the

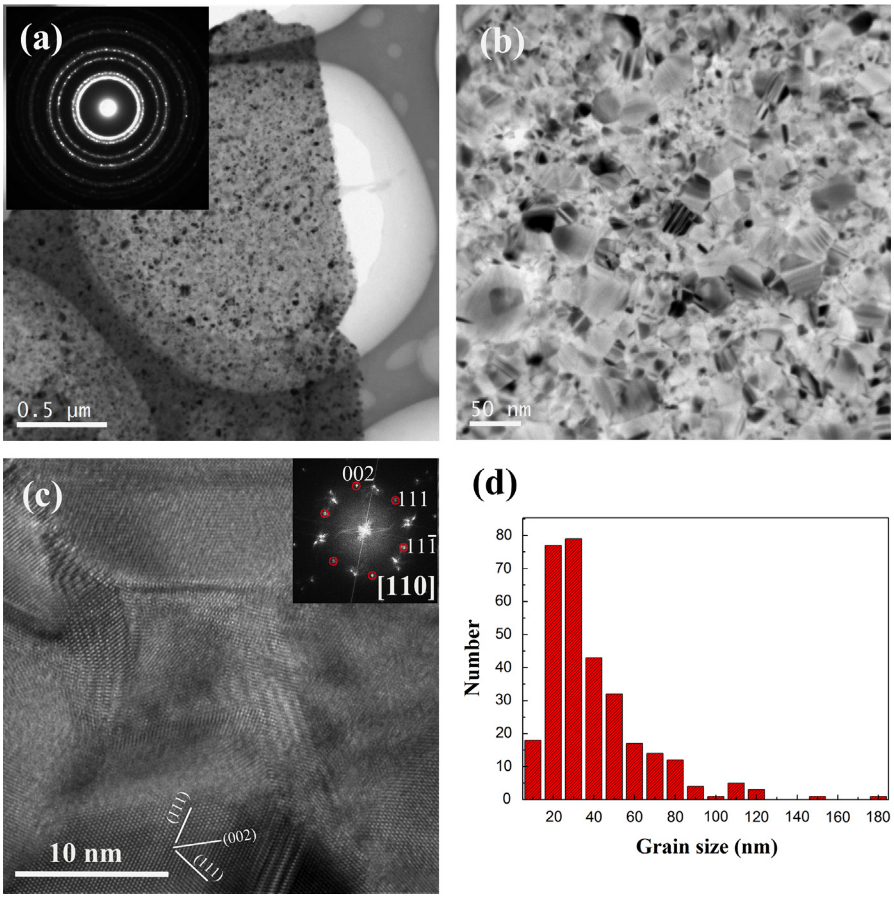

**Fig. 1.** The microstructures of the as-deposited Ag thin films. (a) Low-magnification TEM image of the film. The inset in the upper-left corner is selected area diffraction pattern of the film. (b) A bright-field TEM image of the film. (c) A high-resolution TEM image of the film. The insert in the right upper corner is a Fast Fourier Transform (FFT) pattern showing [110] zone axis diffraction characteristic. (d) The statistical distribution of grain sizes of the film.