



Development of residual strains and their relaxation processes in atomically thin layers of core-shell structured nanoparticles

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ARTICLE INFO

Keywords:

Core-shell structure
Nanoparticle
Residual lattice strain
Dislocation
Strain engineering

ABSTRACT

Residual elastic strains imposed on atomically thin films coated onto substrate materials cause changes in the electronic structures of such films. This knowledge has been employed in various industrial sectors as a means of tuning the optical, electrical, magnetic, and chemical properties of materials. Although the magnitudes of residual strains and properties of materials are closely related to the thickness of coating layers, the measurement of residual strains that develop in atomically thin films is challenging owing to the experimental complexities of conventional measurement methods. In this work, the residual strains developed in atomically thin Au overlayers grown epitaxially on Pd nanoparticles were measured using transmission electron microscopy based on nanobeam precession electron diffraction. Experiments revealed that a 1-nm-thick Au film can withstand an exceptionally high compressive strain of 4.3%, which is reduced to nearly zero as the film thickness increases beyond 5 nm. The microstructural evolution of Au films was monitored using high-resolution transmission electron microscopy and the large strain storage capacity and plastic relaxation behavior of Au films were interpreted by comparing them with the previous computer simulations and theoretical models.

1. Introduction

It is well known that the properties of materials are closely related to their structures. Controlling the chemical composition of a material is one of the most common approaches to altering its structure and controlling the concomitant changes in associated properties. Another technique that is used to alter material structure is the imposition of elastic strain on material surfaces [1,2]. The basic principle behind this technique is the induction of large residual elastic strains on atomically thin films coated onto substrate materials in a controlled manner. Residual strain causes changes in the electronic structures of films, thereby altering their surface properties. Since the first successful application of this technique, which is known as “strain engineering,” to semiconductor devices [2,3], it has been applied in various industrial sectors as a means of tuning the optical [4], electrical [5], magnetic [6], and chemical properties [7] of materials. However, as film thickness increases, the stresses developed in a film can promote the local plastic yielding of film materials, which relieves residual strains and leads to an

eventual loss of desired properties. Therefore, measuring the magnitude of strain as a function of film thickness and crystallographic orientation is crucial for establishing the structure-property relationships of thin films coated onto substrate materials.

The residual strains imposed on thin films are typically measured using techniques such as X-ray diffraction (XRD) [8] and X-ray absorption fine-structure spectroscopy [9]. Generally, these methods yield strains as average values between film and substrate materials, which are often difficult to separate. Other techniques based on transmission electron microscopy (TEM), such as geometrical phase analysis [9,10] and dark-field electron holography [11], are also widely used. However, in these methods, the limited field size for obtaining high-resolution images and the experimental difficulty of aligning zone axes hinder the precise determination of strains and their distributions in materials over large areas. Unlike methods based on XRD and conventional TEM, nanobeam precession electron diffraction (NPED) facilitates the acquisition of high-precision strain maps from material regions on the order of a few hundred nanometers, even without alignment to precise zone axes

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