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Nonlinear optical properties of thin iron films grown on MgO (100) by pulsed laser deposition

W.T. Wang[a,b,*], D.Y. Guan[b], G. Yang[b], G.Z. Yang[b], Y.L. Zhou[b], H.B. Lu[b], Z.H. Chen[b]

[a] *Institute of Opto-Electronic Information Science and Technology, Yantai University, Yantai 264005, PR China*
[b] *Laboratory of Optical Physics, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, PR China*

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Abstract

Thin films of iron were fabricated on MgO (100) substrates using pulsed laser deposition technique. The crystalline property and surface roughness of the films were investigated by X-ray diffraction and atomic force microscopy, respectively. It was found that the morphology of the Fe film deposited at room temperature was characteristic of continuous metal film. Increasing the deposition temperature caused the smooth Fe film to break up, and nanoscale Fe islands were formed. We performed the *z*-scan measurements to study the third-order optical nonlinearity of the continuous approximately 9-nm-thick iron film. The results show that the ultrathin iron film exhibits large nonlinear refractive coefficient, $n_2 = 7.09 \times 10^{-5}$ cm^2/kW, and nonlinear absorption coefficient, $\beta = -5.52 \times 10^{-3}$ (cm/W), at the wavelength of 532 nm.

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Keywords: Iron films; Structure and morphology; Optical nonlinearity; Pulsed laser deposition

1. Introduction

Thin iron films deposited on certain substrates like GaAs or MgO have been intensively studied for their fundamental and technological properties. Many works were dedicated to the growth mode, magnetization reversal, or magneto-optic effects of epitaxial Fe films [1–3]. Except for magnetic characters, Fe films also exhibit interesting optical properties [4]. Recently, we have reported that the composite materials containing Fe nanoparticles have large optical nonlinearity [5]. However, the inherent nonlinear optical properties of thin Fe films have not been measured directly.

In the present work, we prepared thin iron films on MgO (100) substrates using pulsed laser deposition (PLD) technique. The effects of deposition temperature on the crystalline structure and surface morphology of Fe films were investigated by X-ray diffraction (XRD) and atomic force microscopy (AFM). We examined, for the first time to our knowledge, the inherent nonlinearities of iron metal directly using the *z*-scan method [6,7].

2. Experimental details

The films were deposited in vacuum (higher than 5×10^{-4} Pa) onto MgO (100) substrates which were polished on both sides (0.5 mm in thickness). A Lambda Physic XeCl excimer laser (308 nm, 20 ns full width at half maximum) operating at 4 Hz repetition rate was focused onto a high-purity (99.999%) target of iron. The typical energy density at the target surface was closed to 2 J/cm^2, high enough to ablate the metal. The target was mounted on a rotating holder, 35 mm from the MgO (100) substrates. All the MgO substrates were first cleaned in an acetone ultrasonic bath before being placed

* Corresponding author. Laboratory of Optical Physics, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, PR China. Tel./fax: +86-1082-6492-99.
E-mail address: wwt@aphy.iphy.ac.cn (W.T. Wang).

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into the deposition chamber. Three samples were prepared at room temperature, 500 °C, and 800 °C, respectively. The thickness of the samples was measured to be approximately 110 nm. The surface morphology was investigated by AFM (Digital Instrument Nanoscope IIIa) in contact mode with a NPS-type Si_3N_4 tip. A $\theta-2\theta$ scan with Cu Kα radiation at 1.54Å was used to determine the crystallinity. A VGESCALab-5 X-ray photoelectron spectroscopy (XPS) was used to detect the chemical nature of the Fe films. XPS spectra were measured under a vacuum of 1.33×10^{-8} Pa using Mg Kα exciting radiation (1253.6 eV).

Thick Fe films have low transmittance values that are not suitable for the measurements of optical nonlinearity. Ultrathin Fe films must be prepared in order to obtain a significant signal from the *z*-scan. For this purpose, we fabricated another sample for the *z*-scan measurements, which was prepared under the same experimental conditions as that of the sample prepared at room temperature, except that the deposition time was only 2 min, and the thickness of this sample was determined to be about 9 nm. A 1-nm-thick layer of Au was deposited on this sample subsequently to protect the Fe films from oxidation.

The optical properties of the ultrathin Fe film were studied by the *z*-scan method, which is one of the most successful techniques for measuring the third-order nonlinearity. During the *z*-scan measurements, the sample is moved along the *z*-axis of a focused Gaussian-profile laser beam, the power of which is kept constant during the whole scan. If part of the light intensity transmitted across the nonlinear material is measured through an aperture in front of the detector, the magnitude and the sign of the nonlinear refractive index n_2 is obtained. In this case, it refers to a closed-aperture (CA) *z*-scan. If all the transmitted light is detected by takeoff from the aperture, the nonlinear absorption is manifested on the so-called open-aperture (OA) *z*-scan. The nonlinear refractive index (n_2) and the nonlinear absorption coefficient (β) are defined by

$$n(I) = n_0 + n_2 I,$$

and

$$\alpha(I) = \alpha_0 + \beta I,$$

respectively, where n_0 is the linear refraction index, α_0 is the linear absorption coefficient, and I denotes the beam intensity.

In our experiment, a Q-switched Nd:YAG laser frequency doubled at 532 nm and characterized by a pulse duration of 10 ns at a repetition rate of 1 Hz was employed as the light source. The laser beam was focused on the sample with a 120 mm focal length lens, leading to a measured beam waist of 30 μm and a pulse energy of 11 μJ in the focal plane. To use the *z*-scan technique correctly, experimental conditions were carefully controlled. Firstly, we used the two identical photodetectors and an energy ratiometer to correct the raw data. The effects of the fluctuations from the laser power were eliminated by dividing the transmitted power by the power at the reference detector. Secondly, we set the repetition rate to 1 Hz in order to reduce the possible thermal accumulative effect. Finally, the transmitted energy was measured and a transmission of the sample was calculated by averaging the data coming from 100 laser pulses. In order to check the uniformity of the samples, the measurement was repeated on the same spot or on different spots of the same sample. The same results were obtained under the same measurement conditions.

3. Results and discussion

Fig. 1 shows 5×5 μm^2 AFM images of the samples prepared at room temperature, 500 °C, and 800 °C. It is clear from Fig. 1a that a continuous smooth iron film can be formed when the substrate was kept at room temperature during the entire deposition process. The root-mean-square surface roughness is only 0.380 nm, suggesting excellent smoothness and uniformity of the Fe film. However, a distinct transformation in the surface morphology has been observed as presented in Fig. 1b and (c) for the growth temperature of 500 and 800 °C, respectively. The image of the sample prepared at 500 °C shows large islands of widely varying shapes and sizes with square terraces on the surface. When the deposition temperature is increased to 800 °C, the very tall islands come into being, and some smaller islands are observable between them. It seems likely that with the increasing of deposition temperature, the continuous Fe films break up and nanoscale Fe islands grow on the surface, and the smoothness of the films is degenerated.

Fig. 2 shows the XRD patterns of the samples. Film grown at room temperature shows no evident diffraction peak corresponding to iron (Fig. 2a), indicating that the amorphous Fe film is formed. When the substrate is kept at 500 °C during Fe deposition, the XRD spectra show a preferential (200) orientation which also coincides with the so called [8] easy magnetization family of planes (Fig. 2b). This observation is repeated at 800 °C with increased intensity of Fe (200) peak (Fig. 2c). It is clear that the Fe films deposited at 500 and 800 °C are of single phase, and no diffraction from randomly oriented grains or impurity phases can be observed from the XRD patterns. The growth mode was that the (100) Fe films ($a_{\text{Fe-bulk}}$ = 2.866 Å) grew epitaxially in the BCC phase on the (100) MgO substrate (a_{MgO} = 4.213 Å), with 45 in-plane rotation of the structure, which is the same as reported previously [2].