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

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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 $5 \times 10^{-4}$ 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

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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 $1.33\times10^{-8}$ 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 ($\beta$) 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, $\alpha_0$ 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 $5\times5$ 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 ($a_{\text{MgO}}$ = 4.213 Å), with 45 in-plane rotation of the structure, which is the same as reported previously [2].

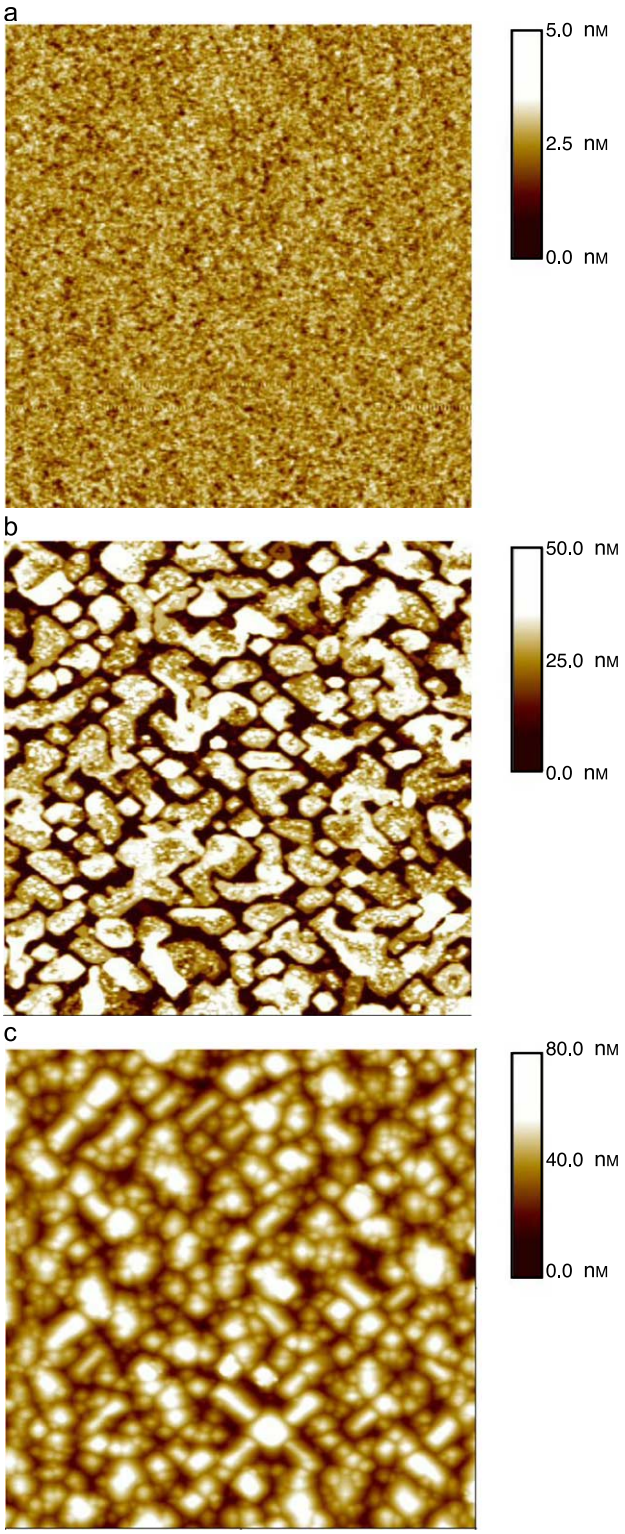


Fig. 1. AFM images of the samples prepared at (a) room temperature, (b) 500 °C, and (c) 800 °C.

XPS spectra of the sample prepared at room temperature are shown in Fig. 3. The binding energies were corrected with reference to the assumed value of 284.6 eV for the resulting C 1s line from the adsorbed hydrocarbon contaminant. Fig. 3a exhibits the chemical states of iron on the surface of the film. It is evident that the Fe $2p_{3/2}$ signal splits into two peaks, which corresponds to pure iron (707.1 eV) and $Fe_2O_3$ (710.8 eV), respectively. However, when the film was bombarded by an argon ion beam (2.5 kV, 20 μA) for 2 min, the signal of Fe in oxidized state disappeared as shown in Fig. 3b. We also measured the chemical nature of the samples prepared at 500 and 800 °C, and obtained the similar results. These XPS results suggest that the surface of Fe films is easy to be oxidized in air whether the films were deposited at room temperature or at high temperature, but the thickness of the iron oxide was about 2 nm.

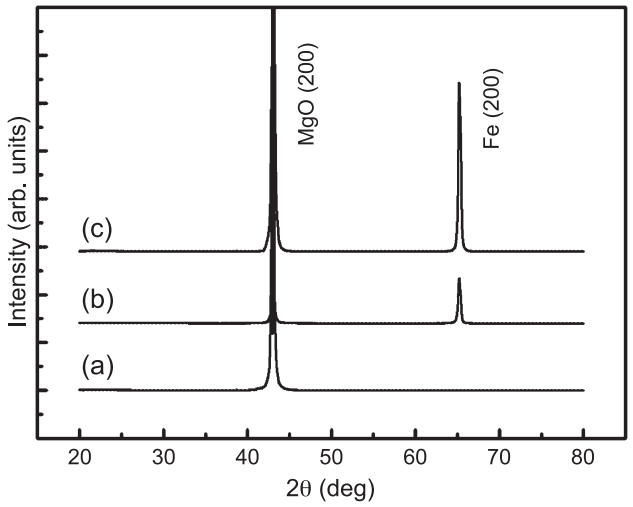


Fig. 2. XRD patterns of the samples prepared at (a) room temperature, (b) 500 °C, and (c) 800 °C.

We used the 9-nm-thick room-temperature-grown sample for the inherent nonlinearity measurements of iron metal by the *z*-scan method. The CA and OA results are

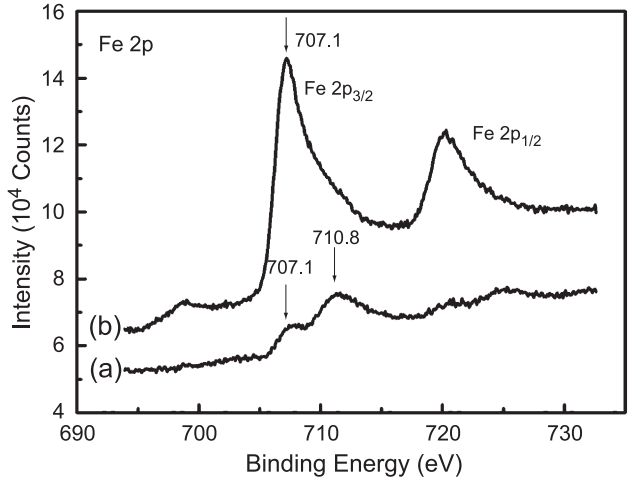


Fig. 3. Fe 2p XPS spectra of the sample prepared at room temperature: (a) surface of the film, and (b) being etched by an $Ar^+$ ion beam (2.5 kV, 20 μA) for 2 min.

shown in Fig. 4a and b, respectively. The CA curve has a valley-peak configuration, corresponding to a positive nonlinear refractive index. The OA curve comprises a normalized transmittance peak, indicating the presence of nonlinear saturable absorption. Because the MgO substrate in our experiment has weak nonlinear optical response at 532 nm, which has been measured by the same method, the high nonlinear optical properties observed here result from the Fe film.

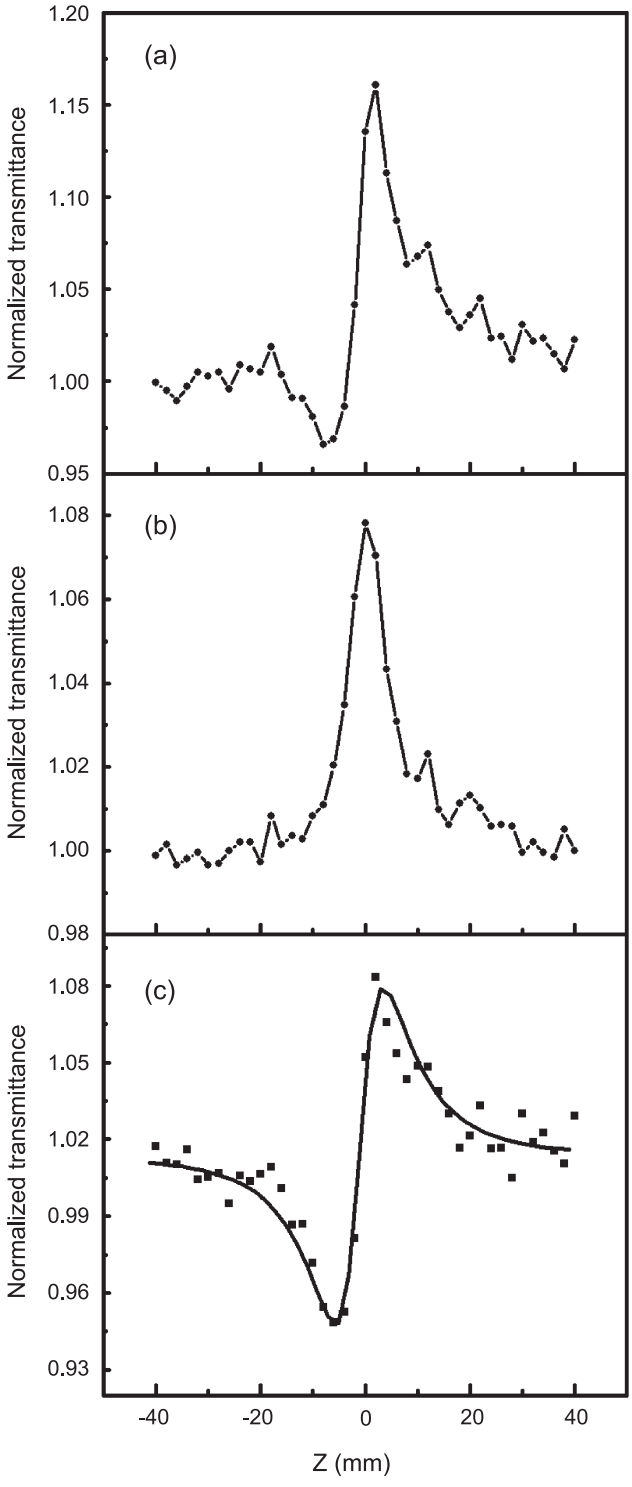


Fig. 4. *Z*-scan normalized transmittance with (a) CA and (b) OA of the 9 nm-thick room-temperature-grown sample. (c) Normalized results (filled squares) for the data in (a) divided by those in (b), and the solid line is the theoretical fit to the data.

The calculations of the values of $n_2$ and $\beta$ have been discussed in detail elsewhere [9–11]. The absorption coefficient $\beta$ (m/W) can be calculated from the OA normalized transmittance in Fig. 4b. The obtained $\beta$ (m/W) of the Fe film is $-5.52\times10^{-3}$ (cm/W). In order to obtain quantitatively the nonlinear refractive coefficient shown in Fig. 4a, a simple and approximate method [6] was used, in which the CA data were divided by the corresponding OA data, and the results were shown in Fig. 4c. The calculated $n_2$ of the Fe film is $7.09\times10^{-5}$ cm$^2$/kW. The uncertainty in the obtained values arises essentially from those in the fluence values, i.e., beam waist, pulse width, and energy calibration. The relative uncertainty in the values is estimated to be 10–20%, as is typical of *z*-scan measurements. It is worth noting that the obtained values of $\beta$ and $n_2$ were representative of bulk Fe material because the sample was a continuous iron thin film.

The obtained values of $n_2$ and $\beta$ of iron film are comparable with those of Au films [12]. However, the optical nonlinearity of thin iron films is larger than that of the composite films containing Fe nanoparticles [5]. As is known, the measurements of the third-order optical nonlinearity of a material are sensitive to the character of the used laser beam [13]. When a nanosecond laser is used, as in most of the early experiments [14], the thermal effect arises, which can enhance the optical nonlinearity. In our experiments, the linear absorption of the thin Fe film is larger than that of composite film containing Fe nanoparticles at 532 nm. As a consequence, the thermo-optic effect (nonlinear thermal lensing) causes an increase in the nonlinear refraction and absorption of the thin Fe film as the ns laser pulse heats it. Moreover, the layer of Au on the surface of Fe film can also contribute to the large observed optical nonlinearity since the nonlinear absorption of a thin gold film at 532 nm has been reported [12].

## 4. Conclusion

In summary, thin Fe films grown on MgO substrates were prepared by PLD. The crystalline property and surface morphology of the films deposited at different substrate temperatures were investigated. The inherent nonlinear optical properties were determined through direct measurements on the continuous ultrathin Fe film. The *z*-scan results indicated that the Fe films had large nonlinear refractive coefficient, $n_2=7.09\times10^{-5}$ cm$^2$/kW, and nonlinear absorption coefficient, $\beta=-5.52\times10^{-3}$ (cm/W).

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