Magnetic properties of nanocrystalline Fe$_3$O$_4$ films

Jinke Tang  
*University of Wyoming, jtang2@uwyo.edu*

Kai-Ying Wang  
*University of New Orleans*

Weilie Zhou  
*University of New Orleans*

Follow this and additional works at: [https://repository.uwyo.edu/physics_astronomy_facpub](https://repository.uwyo.edu/physics_astronomy_facpub)

Part of the Physical Sciences and Mathematics Commons

Publication Information

This Article is brought to you for free and open access by the Physics and Astronomy at Wyoming Scholars Repository. It has been accepted for inclusion in Physics and Astronomy Faculty Publications by an authorized administrator of Wyoming Scholars Repository. For more information, please contact scholcom@uwyo.edu.
Magnetic properties of nanocrystalline Fe$_3$O$_4$ films
Jinke Tang, Kai-Ying Wang, and Weilie Zhou

Citation: Journal of Applied Physics 89, 7690 (2001); doi: 10.1063/1.1358350
View online: http://dx.doi.org/10.1063/1.1358350
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/89/11?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Effect of silver addition on structural, electrical and magnetic properties of Fe$_3$O$_4$ thin films prepared by pulsed laser deposition
J. Appl. Phys. 111, 073907 (2012); 10.1063/1.3702463

Granular growth of Fe$_3$O$_4$ thin films and its antiphase boundaries prepared by pulsed laser deposition
J. Appl. Phys. 89, 7398 (2001); 10.1063/1.1358831

Magnetic and magneto-transport properties of γ Fe$_2$O$_3$-grain-embedded Fe$_3$O$_4$ thin films

An effect of nitrogen on magnetic properties and microstructure of Fe–Nb–B–N nanocrystalline thin films

Magnetic properties of sputtered Fe$_{1-x}$O and Fe+Fe$_3$O$_4$ thin films
J. Appl. Phys. 81, 5250 (1997); 10.1063/1.364483
Magnetic properties of nanocrystalline \( \text{Fe}_3\text{O}_4 \) films

Jinke Tang
Department of Physics, University of New Orleans, New Orleans, Louisiana 70148

Kai-Ying Wang and Weilie Zhou
Advanced Materials Research Institute, University of New Orleans, New Orleans, Louisiana 70148

Nanocrystalline magnetite \( \text{Fe}_3\text{O}_4 \) films of about 180 nm thick have been deposited on Si(100) substrates by pulsed laser deposition. Zero-field-cooled magnetization shows clearly the Verwey transition near 120 K by an abrupt change, which is absent from the field-cooled magnetization. This is correlated to its hysteresis curves where the loops remain open until a high field of 2 T. The magnetization does not saturate in field 2 orders of magnitude higher than its coercive field. Such behaviors may result from the existence of antiphase domains. Antiphase boundaries inside the grains are clearly observed with transmission electron microscopy. Negative magnetoresistance of about 12% has been observed near 120 K in a field of 9 T. © 2001 American Institute of Physics.

Pulsed laser deposition (PLD) is an attractive method for preparation of a variety of films.\(^1\) It is a highly versatile tool for a range of thin films from nanocomposite to epitaxial growth.\(^2\) A few of the characteristic features of PLD are stoichiometric transfer, growth from an energetic beam, reactive deposition, and its simplicity to operate. Highly nanocrystalline or cluster-assembled films have been produced by PLD. PLD has also been used to form other exotic hybrid or crystalline or cluster-assembled films have been produced by PLD. PLD has also been used to form other exotic hybrid or composite thin films containing normally incompatible materials that could be difficult to synthesize otherwise.\(^3\)

There has been increased interest in the half-metallic magnetite \( \text{Fe}_3\text{O}_4 \) as its highly spin polarized nature (supposedly \( \sim 100\% \)) is desirable for tunneling magnetoresistance based device applications.\(^4\) PLD has been successfully used recently to grow \( \text{Fe}_3\text{O}_4 \) films by several groups. Epitaxial \( \text{Fe}_3\text{O}_4 \) films were prepared on MgO,\(^5\) SrTiO\(_3\), and \( \alpha\)-Al\(_2\)O\(_3\) substrates by the technique. Polycrystalline \( \text{Fe}_3\text{O}_4 \) films have also been prepared using PLD\(^7\) and other techniques, for example, reactive dc sputtering on Si substrate.\(^8\) The average grain size of these polycrystalline films is about a few micrometers.

Another interesting aspect of thin film \( \text{Fe}_3\text{O}_4 \) is that the magnetization does not saturate in fields 2 orders of magnitude higher than its bulk anisotropy field.\(^9\) This may be due to the presence of antiphase boundaries in single crystal films, across which the exchange coupling is altered.\(^10\) So far, antiphase boundaries have been observed in sputtered and MBE epitaxial \( \text{Fe}_3\text{O}_4 \) films grown on MgO substrates only.\(^11\)

We have prepared nanocrystalline \( \text{Fe}_3\text{O}_4 \) films of average grain size 50 nm on Si substrates by PLD and report in this article some of the important features found in the films. \( \text{Fe}_3\text{O}_4 \) films of about 180 nm thick were deposited on Si (100) substrates by PLD. The \( \alpha\)-Fe\(_2\)O\(_3\) target used for laser ablation was prepared by pressing high purity \( \alpha\)-Fe\(_2\)O\(_3\) powders (99.998%) into a pellet and sintered at 1000 °C for 2 h.

A focused beam of KrF excimer laser (\( \lambda = 248 \) nm) was used for the deposition. The repetition rate was 12 Hz and the Si substrate was heated to 350 °C. Prior to the deposition, an unfocused laser beam was rastered across the Si substrate in a vacuum of \( 3 \times 10^{-6} \) Torr to clean the substrate surface.

Glancing angle x-ray diffraction data were collected with a Philips X'Pert diffractometer using Cu \( K\alpha \) radiation. Figure 1 shows the diffraction pattern of the \( \text{Fe}_3\text{O}_4 \) film. The lattice constant \( a = 0.8392(2) \) nm is close to the powder diffraction (Card No. 19-629) value. Transmission electron microscopy (TEM) was done with a JEOL Model 2010 TEM. Figure 2(top) is a plane-view TEM image of the \( \text{Fe}_3\text{O}_4 \) film. The average grain size is 50 nm. This is much smaller than those in the polycrystalline films mentioned earlier, which is in micrometers.

It is evident from the micrograph that within each single crystal grain it is not uniform. This is due to the presence of antiphase domains in the PLD nanocrystalline \( \text{Fe}_3\text{O}_4 \) films. The TEM image made using (220) reflection shows antiphase domains within each single crystal grain, see Fig.

\(^{a}\)Author to whom correspondence should be addressed; electronic mail: jtang@uno.edu

\(^{b}\) DOI: 10.1063/1.1358350

FIG. 1. Glancing angle x-ray diffraction pattern of a nanocrystalline \( \text{Fe}_3\text{O}_4 \) film.
Our results suggest the antiphase domains also exist in Fe$_3$O$_4$ films deposited on Si substrates, not exclusively on MgO substrates as originally thought.

Magnetization data obtained with a Quantum Design superconducting quantum interference device is shown in Fig. 3 as a function of temperature. A magnetic field of 500 Oe was used for both the zero-field-cooled (ZFC) and field-cooled (FC) measurements. ZFC data clearly show the Verwey transition at about 120 K with the characteristic sharp drop. One of the unique features of the PLD nanocrystalline films is the absence of the sharp drop in the FC data below the Verwey transition in a field of 500 Oe. An abrupt drop is usually observed in FC runs in applied fields up to several kOe in bulk and epitaxial films. We will discuss this feature in conjunction with the magnetic hysteresis curves later.

The temperature dependence of the coercivity $H_c$ and remanence $M_r$ of the Fe$_3$O$_4$ nanocrystalline films is shown in Fig. 4. The remanence increases from 140 to 195 emu/cm$^3$ as temperature decreases from 300 to 5 K. A plateau forms just below the Verwey transition which is consistent with the single crystal data. The coercivity is 275 Oe at 300 K and increases very slowly from 300 to 120 K. Then it undergoes a rapid increase at the Verwey transition and reaches 750 Oe at 5 K. The abrupt increase in coercivity is associated with a change of anisotropy due to the Verwey transition where a structural change from cubic to monoclinic occurs.

As mentioned earlier, the FC magnetization shows no anomaly at the Verwey transition in a field of 500 Oe. This is consistent with the hysteresis data shown in Fig. 5, inset, where the three curves measured at 100, 120, and 140 K,

Strain, size effects, and possible small departure from the precise Fe$_3$O$_4$ stoichiometry might be responsible for the observed broadening.

The temperature dependence of the coercivity $H_c$ and remanence $M_r$ of the Fe$_3$O$_4$ nanocrystalline films is shown in Fig. 4. The remanence increases from 140 to 195 emu/cm$^3$ as temperature decreases from 300 to 5 K. A plateau forms just below the Verwey transition which is consistent with the single crystal data. The coercivity is 275 Oe at 300 K and increases very slowly from 300 to 120 K. Then it undergoes a rapid increase at the Verwey transition and reaches 750 Oe at 5 K. The abrupt increase in coercivity is associated with a change of anisotropy due to the Verwey transition where a structural change from cubic to monoclinic occurs.

As mentioned earlier, the FC magnetization shows no anomaly at the Verwey transition in a field of 500 Oe. This is consistent with the hysteresis data shown in Fig. 5, inset, where the three curves measured at 100, 120, and 140 K,
The grain sizes of the sputtered film were about 1–2 μm. The MR (~1.0% at room temperature and 0.5 T) was attributed to the intergranular transport of spin polarized electrons. On the other hand, comparison between epitaxial films and polycrystalline films prepared by reactive dc sputtering suggests minimal contribution from the electron transport across grain boundaries. Our study does not address specifically whether the spin-dependent intergranular transport plays a dominant role in the observed negative MR, however the connection, if any, between the MR and possible existence of antiphase domains should be explored. It has been argued that the antiphase boundaries may become immobile magnetic domain walls. It is of interest to study the MR due to pinned domain walls as their thickness can become smaller than the spin diffusion length and give large MR.

This research was supported by the Louisiana Board of Regents Support Fund (No. LEQSF(2000-03)-RD-B-10) and Sharp Laboratories of America.