Optical and magnetic studies of NdFeB films

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The optical, electrical, and magnetic properties of magnetic $Nd_{15}Fe_{77}B_8$ and nonmagnetic $Nd_5Fe_{62}B_{33}$ films have been investigated. It has been found that within the visible (wavelengths 4400–6400 Å) the films are transparent with thickness less than 800 Å for magnetic films and 1500 Å for nonmagnetic films. This is due to the difference of the magnetic permeability μ between films. The oscillatory behavior with thickness of the films is observed in the film samples. The anomalous behaviors near the Curie temperature for both bulk- and film-type samples are similar to that of the type-III ferromagnets. The temperature at which the electrical resistance of the film-type samples increases abruptly varies from 620 K for 1000 Å to 650 K for 2000 Å.

I. INTRODUCTION

Permanent magnetic materials based on rare earths and iron have been extensively studied during the past years.[1-3] Many investigations on the relation between the crystal structure and the magnetic properties of the bulk NdFeB system have been reported.[3-6] Comparatively little effort has been devoted to the physical properties of the film-type NdFeB samples. Therefore, it is very interesting to study the properties of film-type permanent magnetic materials. In earlier reports,[7,8] we have investigated the specific heat, electrical resistivity, and magnetization characteristics of the bulk-type NdFeB samples.

In this article we report the optical and magnetic properties of NdFeB films as well as the comparison of the electrical resistivity and magnetization behaviors between the film- and bulk-type NdFeB samples.

II. EXPERIMENT

Two NdFeB bulk ingots with different composition, $Nd_{15}Fe_{77}B_8$ (magnetic) and $Nd_5Fe_{62}B_{33}$ (nonmagnetic), were prepared from commercially available elements of high-purity Nd (99.9%), Fe (99.99%), and B (99.9%). Three different types of samples were prepared; i.e., the arc-melted, sintered, and thin-film types. The arc-melted samples were atomatically weighted according to the compositions of $Nd_{15}Fe_{77}B_8$ and $Nd_5Fe_{62}B_{33}$. Both the chemical analyses and powder-x-ray-diffraction techniques were used to confirm the composition and structure of the NdFeB samples. For preparing the sintered NdFeB samples, the arc-melted ingots were crushed and pulverized to powders with a particle size of roughly 5–10 μm. These powders were pressed in a magnetic field of 2 T and then sintered and annealed in Ar gas between 1370 and 870 K.

The film-type samples were grown in a 10^{-5} Torr vacuum chamber by the electron-bombardment heating (*e*-gun) technique. The arc-melted ingot was contained in a crucible of electrically conducting material as the evaporant, and was bombarded with a beam of electrons to heat and vaporize it. A polished MgO(100) substrate was mounted on a manipulator. The thickness of the films was detected by a crystal thickness monitor. The electrical resistance measurements were performed by using a four-probe method. Both a superconducting quantum interference device (SQUID) and vibrating sample magnetometer were used to determine the magnetization of the samples. The optical transmission and reflection were measured by means of a spectrophotometer. The incident wavelengths for this spectrophotometer are between 2000 and 8500 Å; the incident angle for both transmission and reflection is fixed at 0° and 12°, respectively.

III. RESULTS AND DISCUSSION

The electrical resistivity ρ of the $Nd_{15}Fe_{77}B_8$ and $Nd_5Fe_{62}B_{33}$ bulk samples between 4 and 1200 K has been reported before.[7,8] For the bulk $Nd_{15}Fe_{77}B_8$ sample, we have found that, in the vicinity of the Curie temperature T_c, the temperature coefficient of the electrical resistivity $d\rho/dT$ decreases before T_c and abruptly increases after T_c as the temperature increases. We call this behavior the inverse λ-type anomaly. This is completely different from the λ-type anomaly near the Curie temperature as shown in the nickel-like ferromagnet;[9] but, it is quite similar to that of the type-III ferromagnet.[10] Figure 1 shows the electrical resistivity R and its temperature coefficient dR/dT of a film-type $Nd_{15}Fe_{77}B_8$ sample with the thickness of 1000 Å as functions of temperature between 570 and 620 K. Clearly, it shows similar behavior as the bulk samples, except that T_c is slightly shifted to a higher temperature. The origin of this shift is not well understood at the present time. For the nonmagnetic $Nd_5Fe_{62}B_{33}$ sample, both the

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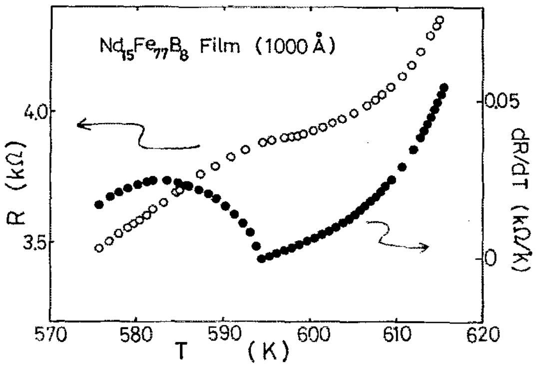

FIG. 1. R and dR/dT as functions of temperatures between 570 and 620 K for $Nd_{15}Fe_{77}B_8$ film with thickness of 1000 Å.

electrical resistance and its temperature coefficient are monotonically varied functions of the temperature; no anomaly was observed. Therefore, we conclude that only for the magnetic $Nd_{15}Fe_{77}B_8$ samples (bulk and film type) does the electrical resistivity in the vicinity of T_c behave similarly to that of the type-III ferromagnet.

We have measured the electrical resistance of the film-type NdFeB samples with the thickness varied between 1000 and 2000 Å for temperatures below 700 K. Besides the anomaly near T_c for $Nd_{15}Fe_{77}B_8$ which has been discussed above, another anomalous behavior for all the film-type samples is that the resistance increases abruptly with increasing temperature after a critical temperature T_x. As an example, Fig. 2 shows the R/R_0 as a function of temperature between 460 and 660 K for two samples: $Nd_{15}Fe_{77}B_8$ (thickness 1000 Å) and $Nd_5Fe_{62}B_{33}$ (thickness 2000 Å). R_0 is the electrical resistance at 460 K for each sample. Generally speaking, T_x for all 1000 Å samples is lower than that of 2000 Å samples. This anomaly could be due to the following two possibilities: The first possibility could be that the reaction between the film and the MgO substrate occurs above T_x; the second reason may be due to the evaporation of the film-type sample. Further experimental data are needed for identification.

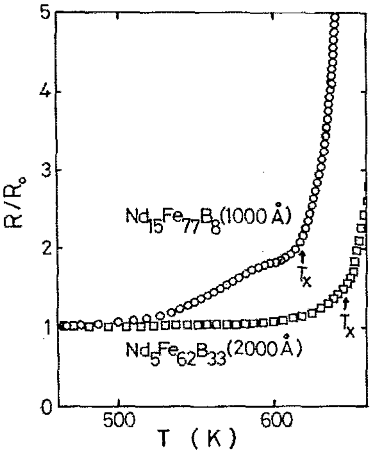

FIG. 2. Normalized electrical resistance R/R_0 as a function of temperature for $Nd_{15}Fe_{77}B_8$ film (thickness of 1000 Å) and $Nd_5Fe_{62}B_{33}$ film (thickness of 2000 Å). R_0 is the electrical resistance at 460 K for each sample.

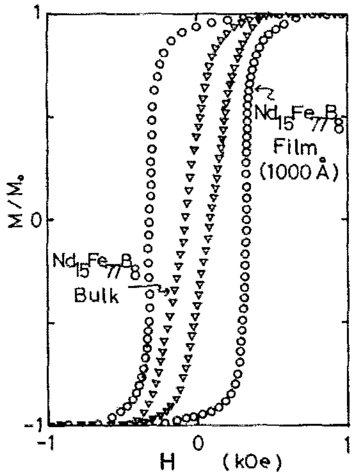

FIG. 3. Normalized magnetization M/M_0 as a function of magnetic field H for film-type and arc-melted $Nd_{15}Fe_{77}B_8$ samples. M_0 is the magnetization at 1000 Oe for each sample.

Figure 3 presents the comparison of the normalized magnetizations between the film-type $Nd_{15}Fe_{77}B_8$ sample with thickness of 1000 Å and the arc-melted sample which was used as the target for preparing this film sample. Here, we normalized the magnetization with the reading at 1000 Oe. It is clear that the coercivity increases after evaporating a nonmagnetized $Nd_{15}Fe_{77}B_8$ ingot to a film type. This means that the anisotropy of magnetization can increase for film-type samples. A permanent magnetic material must have reasonable uniaxial anisotropy for high coercivity. The $Nd_2Fe_{14}B$ compounds are tetragonal in structure[4] with the easy magnetization in the c-axis direction.[11] The best magnetic properties of the sintered samples in this investigation are with an intrinsic coercivity of 14 kOe and an energy product of 27 MG Oe. However, for the film-type NdFeB samples up to now, we have not yet obtained a high enough value of the coercivity (<400 Oe). Further research work to find a film with an epitaxial growth is definitely needed.

The transmittance Tr (or reflectance R) of a film is defined as the ratio of the transmitted (or reflected) energy to the incident energy. Usually, a metal film is an absorbing medium; and the absorptance A of a metal film is connected with R and Tr by the relationship $R+Tr+A=1$.

Figures 4 and 5 shows the Tr and R of $Nd_{15}F_{77}B_8$ and $Nd_5Fe_{62}B_{33}$ film samples as functions of the film thickness with incident wave length at 4400, 5400, and 6400 Å. Clearly, from Fig. 4, the value of the Tr decreases as the thickness of the films increases; and it goes to zero roughly at 800 Å for magnetic $Nd_{15}Fe_{77}B_8$ films and 1500 Å for nonmagnetic $Nd_5Fe_{62}B_{33}$ films. At any specific film thickness below 800 Å, the transmission of nonmagnetic NdFeB metal films is always larger than that of a magnetic NdFeB metal films. On the contrary, from Fig. 5 it is observed that

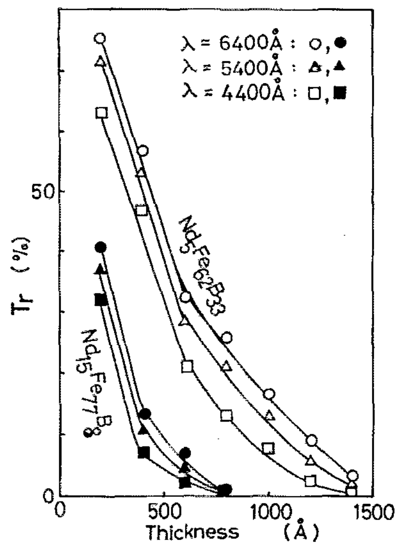

FIG. 4. The transmittance as functions of the film thickness for $Nd_{15}Fe_{77}B_8$ and $Nd_5Fe_{62}B_{33}$ films with incident wavelengths of 4400, 5400, and 6400 Å.

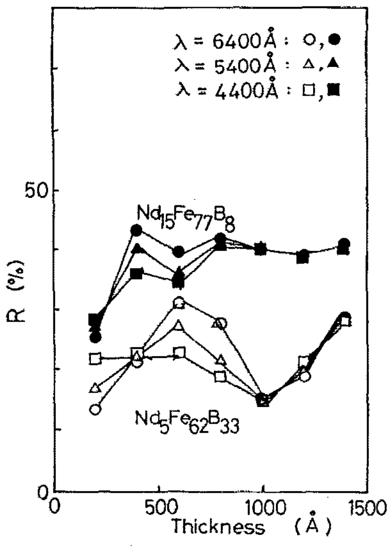

FIG. 5. The reflectance as functions of the film thickness for $Nd_{15}Fe_{77}B_8$ and $Nd_5Fe_{62}B_{33}$ films with incident wavelengths of 4400, 5400, and 6400 Å.

the value of R of the nonmagnetic films is always smaller than that of magnetic films. The oscillatory behavior of R we observed for both films is qualitatively consistent with that of the theoretical variation of R from an absorbing surface.[12]

From the experimental data of R and Tr, we can easily calculate the real part n and the imaginary part k of the complex refractive index $(n-ik)$ for thin films. As an example, for films (thickness of 400 Å) with incident wavelengths of 6400 Å, we obtained $n=0.5$ and $k=1.3$ for nonmagnetic $Nd_5Fe_{62}B_{33}$ films and $n=1.7$ and $k=2.3$ for magnetic $Nd_{15}Fe_{77}B_8$ films. This means that both n and k for magnetic films are larger than those of nonmagnetic films. This can be qualitatively understood by considering the propagation of a uniform plane electromagnetic wave in a conductor. According to the electromagnetic theory in a good conductor,[13] both n and k are proportional to the square root of the relative magnetic permeability μ. In this study, all the NdFeB samples are good conductors. We know that μ of the magnetic $N_{15}Fe_{77}B_8$ samples should be larger than that of the nonmagnetic $Nd_5Fe_{62}B_{33}$ samples. Therefore, our experimental results are qualitatively explained by the basic electromagnetic theory. Further quantitative analyses of our results with the electromagnetic theories are in progress now and will be reported later.

ACKNOWLEDGMENTS

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