Magnetization Study in Fe3O4/MgO Superlattices

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Abstract--- **Magnetization of a series of Epitaxial Fe_3O_4/MgO multilayers grown on MgO(100) substrates by oxygen-plasma-assisted molecular-beam epitaxy has been studied. The Curie temperature decreases with decreasing the thickness of each Fe_3O_4 layer. The saturation magnetization of all the multilayer samples seems to be lower than that of the 200 nm thick Fe_3O_4 film**

INTRODUCTION

In recent years, due to the fast development of various thin-film growth techniques, there has been increased interest in various properties of multilayer system. By reducing the thickness of each layer in multilayer films, the finite size effect related to the reduction from 3 to 2 dimensions can be studied in this way [1-3]. Although a number of interesting effects have been observed on many magnetic/nonmagnetic multilayers, most of the systems are metallic[4,5]. The insulating systems, such as oxides, have localized electronic states which may be understood more easily[6-8]. In this investigation, we report on the first observation of the finite size effect in a ferrimagnetic/ nonmagnetic oxide system: Fe_3O_4/MgO superlattice grown onto MgO(100) substrate.

EXPERIMENTAL CONSIDERATION

Epitaxial Fe_3O_4/MgO multilayers have been successfully grown on MgO(100) substrates by oxygen-plasma-assisted molecular-beam epitaxy. Fe_3O_4 has a cubic spinel structure (a= 0.8398 nm) and MgO has a NaCl cubic structure (a=0.4212 nm). Small lattice mismatch (~ 0.3%) and chemical resemblance make this system an ideal candidate of superlattice structure[6]. Four superlattice samples were grown with alternating layers of ferrimagnetic Fe_3O_4 and dia-magnetic MgO plus a pure thick Fe_3O_4 film for comparison. The thickness of every MgO layer was kept between 1.9 and 2.4 nm, and the thickness of every Fe_3O_4 layer was varied from 1 to 8.2 nm. The total thickness of the films is around 150-300 nm (30-50 bilayer repeats). The long modulation coherency and sharp interface of the superlattices are evidenced by x-ray diffraction. A representative XRD result including the low and mid angle diffractions for a typical superlattice is shown in Fig. 1. Many order of sidebands are shown which indicates the excellent crystalline structure of this film. Similar XRD results are seen on all of the Fe_3O_4/MgO superlattices. Since the roughness of the interface is sensitive to the magnetic property, especially for short modulation wavelength samples, detailed in-situ studies, including Refection High Energy Electron Diffraction (RHEED) and Atomic Force Microscopy (AFM), are also performed. The AFM result reveals that the surface is much flatter for the superlattice samples relative to the pure films. This smoothing is due to an unique growth mode which only exists in the alternation of the constituients and will be discussed elsewhere.

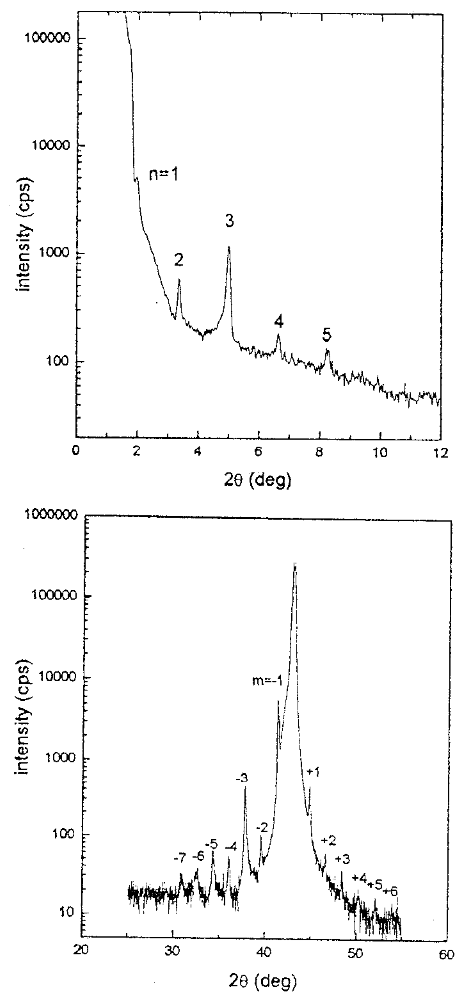

Fig. 1 (a) Low angle and (b) mid angle x-ray diffraction result for a superlattice with $[Fe_3O_4(3nm)/MgO(2.4nm)]_{31.5}$/MgO/MgO(100).

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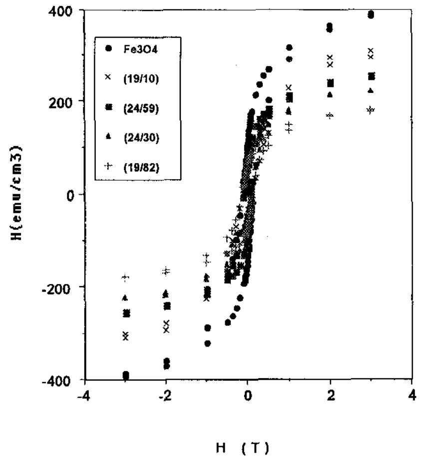

Fig. 2 The magnetization per unit volume of Fe_3O_4 layers for all the samples at T = 5 K as a function of magnetic field between ± 3 T.

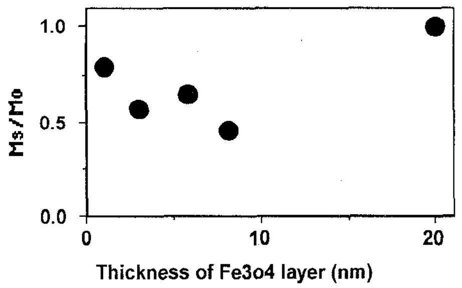

Fig. 3 M_s/M_0 as a function of the thickness of each Fe_3O_4 layer.

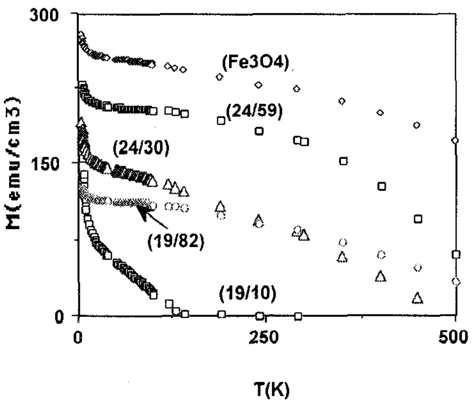

Fig. 4 The magnetization as a function of temperature below 500 K for all the samples with FC and at magnetic field of 0.5 T.

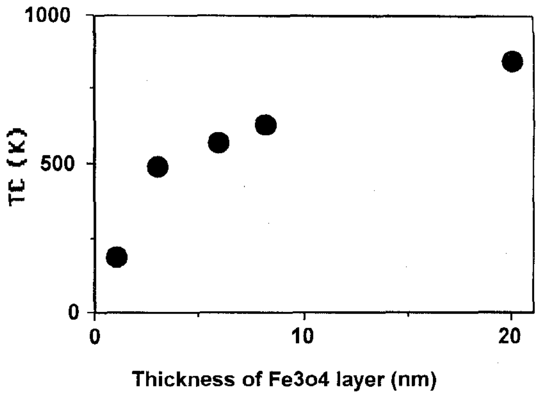

Fig. 5 The Curie temperature as a function of the thickness of each Fe_3O_4 layer.

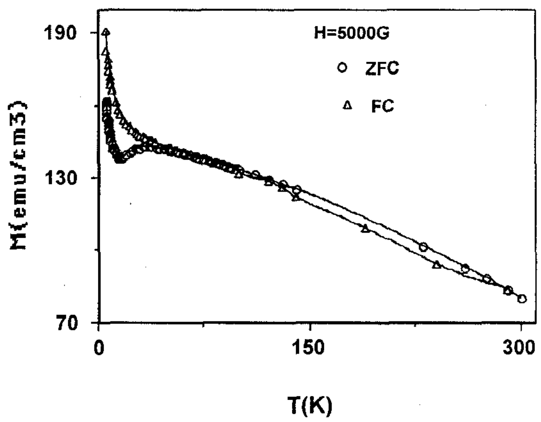

Fig. 6 ZFC and FC magnetization as a function of temperature for $[Fe_3O_4(3nm)/MgO(2.4nm)]_{31.5}$ sample

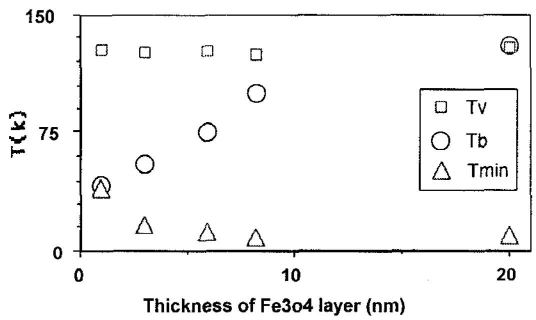

Fig. 7 T_v, T_b, and T_{min} as functions of the thickness of each Fe_3O_4 layer.

The magnetization hysteresis curves were measured using a SQUID magnetometer with the applied field H varied between ±5.5 T. The applied field and the moment direction measured in the most our data were parallel to the superlattice plane (along the crystalline <100> direction). Background Subtraction of the MgO substrate was critical for the accuracy analysis of all samples. The MgO substrates had a very small diamagnetic moment for temperature roughly above 10 K. However, for lower temperatures, it also had a paramagnetic component, which was attributed to the defects in MgO substrates.

RESULTS AND DISCUSSION

Ferrimagnetic/antiferromagnetic superlattices such as Fe_3O_4/NiO and Fe_3O_4/CoO have been studied recently and anomalous magnetic properties have been found[6-9]. These effects typically become more pronounced when the layer thickness gets very thin. Since both materials in these systems have magnetic ordering, the long range correlation may not be cease completely. The effect of exchange coupling between antiferromagnetic and ferro- or ferrimagnetic layers adds extra complexity. Now with Fe_3O_4 separated by a highly insulating nonmagnetic compound of MgO, the finite size effect of Fe_3O_4 can be tested and these results should be useful to interpret other more complicated multilayered systems.

The magnetization hysteresis curves were measured at T = 5 K using a SQUID magnetometer with the applied field H varied between ± 5 T. Background substraction of the MgO substrate was critical for the accuracy analysis of all samples. In general, the magnetization is saturated above 3 T for all the samples. Fig. 2 shows the magnetization per unit volume of Fe_3O_4 layers for four Fe_3O_4/MgO superlattice samples with a thin buffer layer of MgO grown on MgO(100) substracts, {i.e. $[Fe_3O_4(1nm)/MgO(1.9nm)]_{51.5}$, $[Fe_3O_4(3nm)/MgO(2.4nm)]_{31.5}$, $[Fe_3O_4(5.9nm)/MgO(2.4 nm)]_{30}$, $[Fe_3O_4(8.2nm)/MgO(1.9nm)]_{30.5}$, and a pure Fe_3O_4 film with thickness of 200 nm as a function of magnetic field between ± 3 T. It is interested to note that the saturation magnetization behaves as a function of the thickness of each Fe_3O_4 layer as shown in Fig. 3. It is normalized to the saturation magnetization M_0 of the pure Fe_3O_4 film. However, due to an insufficient number of data points, here we only show that the saturation magnetization of all the multilayer samples seems to be lower than that of the 200 nm thick Fe_3O_4 film. Besides, due to that the impurities in the MgO substrates can contribute a significant signal to the saturation magnetization at 5 K, magnetization loops taken above roughly 20 K are in progress and will be reported later.

Fig. 4 shows the magnetization vs temperature studies for all the samples with field cooling (FC) and at an applied magnetic field of 0.5 T below 500 K. We observed that, even the magnetization is frustrated with the thickness of each Fe_3O_4 layer, the Curie temperature T_c determined from the temperatures with roughly zero magnetization (for samples above 500 K, it was roughly determined from the smooth extension of our data points to zero magnetization) for all the samples decreases with decreasing the thickness of each Fe_3O_4 layer as shown in Fig. 5. It is clear that T_c decreases roughly from 445 to 200 K for samples with the thickness of each Fe_3O_4 layer decreasing from 3 to 1 nm.

From the zero field cooling (ZFC) and FC measurement of the magnetization at an applied field of 0.5 T below room temperature, we observed that all the ZFC curves shows a slope change near 120 K, however, it is a smooth curve near 120 K for all the FC curves. As an example, Fig. 6 shows the ZFC and FC curves below 300 K for $[Fe_3O_4(3nm)/MgO(2.4nm)]_{31.5}$ sample. We defined this temperature as the Verwey transition temperature T_v, and found that it is almost independent of the thickness of the Fe_3O_4 layers. The preferred magnetization axis of Fe_3O_4 is <111> above T_V and very nearly <100> below T_V. We determined another two critical temperatures T_b (the blocking temperature) and T_{min} (the minimum temperature of the ZFC curves) experimentally. T_b is obtained from the intersection of the ZF and ZFC M vs T curves, and is found to decrease roughly from 100 to 42 K for film samples with each Fe_3O_4 layer thickness decreased from 8.2 to 1.0 nm as shown in Fig. 7. However T_{min} determined experimentally as shown in Fig. 7 seems to increase with decreasing the thickness of each Fe_3O_4 layer. The influence of the finite size effect on the magnetism has been always a interesting topic in both experiment and theory. When the thickness of each layer in a multilayer system reach to or below a certain length, the associated properties are substantially changed reflecting the lower dimensionality. Here the variation of T_c, T_b, and T_{min} as a function of the thickness of Fe_3O_4 layers shows qualitatively the influence of the finite size effect on the magnetism in Fe_3O_4/MgO multilayer system.

In summary, we have successfully grown epitaxial Fe_3O_4/MgO multilayers on MgO(100) substrates. The Curie temperature decreases with decreasing the thickness of each Fe_3O_4 layer. The saturation magnetization of all the multilayer samples we studied seems to be lower than that of the 200 nm thick Fe_3O_4 film. Further quantitative analyses are in progress now and will be reported later.

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