Magnetic Properties of ‘Sub’ Monolayered Superlattice of Fe/Mg

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Magnetization measurements have been carried out on a ‘sub’ monolayered artificial superstructure film of [Fe1Mg16]470 evaporated on a mylar substrate. An appreciable anisotropy was observed, which shows the easy direction lying out of the film plane at 4.2 K. Although hysteresis appears at this temperature, the Arrott plot obtained from the data at the higher magnetic fields more than 1000 Oe suggests that the magnetic properties of this sample is considerably different from usual ferromagnets.

KEY WORDS: Magnetism/ Fe/Mg superlattice/ Submonolayer/ Arrott plot/

1. INTRODUCTION

Recent development in the fabrication techniques can provide metallic superlattices of tailored modulation not only for the periodicity but also for the composition or the combination of two elements. Among various classes of novel materials Fe/Mg artificial superstructure films (hereafter abbreviated as ASFs) have attracted a special attention due to the fact that Fe and Mg are mutually insoluble in thermal equilibrium state. According to the Mössbauer measurements, a large hyperfine field on Fe nuclei similar to the value in bulk Fe metal was observed in the ASF which consists of monolayered Fe interfaced with thick Mg layers. This fact is just extraordinary because the surface layer of Fe interfaced with many non-magnetic elements more or less loses its magnetic moment. Eventually the monolayered Fe ASF with thick non-magnetic metal layers becomes non-magnetic such as Fe/V ASF. Thus the monolayered Fe/Mg ASF may possess 2-dimensional ferromagnetic features.

Very recently a new result was observed that the hyperfine field on Fe nuclei in an [Fe1Mg16]470 ASF is still 70% of the bulk value at 4.2 K. Note that [Fe1Mg16]470 represents that this ASF consists of 470 bilayers combined with 1 Å Fe and 16 Å Mg. It means that each Fe lattice in this particular ASF should not be extended in ‘Fe’ layers, but nearly a half of the lattice in each layer is occupied by Fe atoms, since the atomic radius of Fe atoms should be nearly 2 Å. Thus we define this sample as a ‘sub’ monolayered superstructure film. Then we raise a fundamental
question on the magnetic state with Fe lattices of the finite size. In this paper we mainly report ferromagnetic properties of \([\text{Fe}_1\text{Mg}_{16}]_{470}\) ASF at low temperatures.

In the next section the magnetization measurements and the results will be described. The third section will be devoted to discussion and conclusion.

2. MAGNETIZATION MEASUREMENTS

Magnetization measurements have been carried out with the vibration magnetometer at Tohoku. This magnetometer was recently upgraded. Now several scans varying either temperature or field are fully controlled by a dedicated microcomputer PC-9801. The output voltage is picked up by coils attached on the pole piece surfaces of the magnet. The hole current, the thermopower of the Au(Fe)-Chromel coupled wire and so forth, are taken into the digital meters. These digital signals are transferred to the data acquisition memories in the microcomputer. Eventually these data are processed by several softwares.

The magnetic field is applied horizontally and both the magnet and the sample can be rotated around vertical axis. In the present set up we use a sample, stacked 100 pieces of \([\text{Fe}_1\text{Mg}_{16}]\) ASF (about 1 cm³) evaporated on the mylar substrate wrapped by a thin aluminum foil. It was placed at the end of the sample holder rod such that the film plane stands vertically. Temperature was controlled within 0.2±0.01 K. Other details of the magnetometer is described in a separate paper.

Figure 1 depicts typical results of the field scan at 4.2 K, in which the field is applied either perpendicular or parallel to the film plane. It is noted that an appreciable hysteresis was observed in both scans as shown in Fig. 1(b). The fact that easy direction is along perpendicular direction to the film plane is consistent with the Mössbauer measurement for \([\text{Fe}_1\text{Mg}_{16}]\) ASF.⁹

Figure 2 shows the results of thermal scan at two different fields applied in both direction parallel and perpendicular to the film plane. At the low field of 50 Oe, a broad peak appears around 20 K besides that spontaneous magnetization quickly diminishes below 10 K.

On the other hand, the magnetization at 1000 Oe behaves differently; it saturates below 20 K and gradually decreases upon heating. Appreciable effect of the anisotropy continuously appears up to 100 K and finally it disappears above then. The magnetization curve always has a convex curvature with respect to the field up to the room temperature.

Arrott plot was obtained from the magnetization data at various temperatures. As is shown in Fig. 3, the curves below 20 K have sharp bends at low \(H/M\) values. Except the unusual portion at low fields and low temperatures, the Arrott plot represents the similar behavior to that of the paramagnetic state above the Curie temperature for normal ferromagnet. It is interesting that the extrapolating point from the data at the higher magnetic field at 4.2 K crosses at a positive value on the abscissa or a negative value on the ordinate, which conflicts to the fact of the spontaneous magnetization observed at this temperature.

The magnetization curves can be fitted to the Langevin function of \(\coth a - 1/a\),
Fig. 1. (a) Magnetization curves for $[\text{Fe}_1 \text{Mg}_{16}]_{470}$ ASF at 4.2K. Solid and dashed lines respectively represent measurements under applied field in perpendicular direction and parallel to the film plane. Fig. 1 (b) Enlarged portion near low magnetic field in the vicinity of zero field.
Fig. 2. (a) Thermal scans under the constant magnetic field of 50 Oe. Solid and dashed lines mean the same as described in Fig. 1. (b) Scans at 1000 Oe.
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Fig. 3. Arrott plot for \([\text{Fe}_1 \text{Mg}_{16}]_{47\circ}\) ASF. The numbers in the figure represent temperatures at each field scan. Field was applied in perpendicular direction to the film plane.

but the fitted values of a \((H/T)\) vary with the temperature. It means that magnetic properties may not be solely interpreted by a simple superparamagnetism.

3. DISCUSSION AND CONCLUSIONS

We have confirmed that a `sub' monolayered Fe in Fe/Mg ASF possesses ferromagnetic properties and the easy direction is out of plane. The features of magnetization curves are divided by two portions as typically shown by the Arrott plot; the ferromagnetic region which disappears around 10 K and paramagnetic regions. This fact indicates that the Fe lattices are distributed in their size. The dramatic decrease of magnetization seems to indicate the characteristic features of the low dimensionality, but further analysis for the magnetization curves is not easy. Thus the detailed discussion on the 2-dimensional behavior in this material will be left in future studies.

The superparamagnetic behavior is also peculiar. Fitting these magnetization curves to the Langevin function without introducing the distribution of the size does not yield the average size throughout all temperatures measured in the present experiments. Therefore we draw a schematic picture of Fe lattices in the `sub' monolayered Fe/Mg ASF as follows. The size of Fe lattices in which all Fe magnetic moments are ferromagnetically aligned are widely distributed. The thermal fluctuation starts at around 10 K and the residual magnetization begins to decrease. Between ferromagnetic clusters, at least partially, weak anti-ferromagnetic couplings exist (up to around 20 K) which might be an interlayer interaction through Mg layers. They may break around at 20 K. At elevated temperatures above these
critical temperatures, the Fe lattices behave all paramagnetic. Mössbauer spectroscopic measurements showed that the critical temperature for this sample is about 35 K; Namely the fluctuation rate of magnetization at above this temperature is faster than the Mössbauer observation time. In order to clarify these circumstances more accurately, we must elucidate the magnetization processes for other specimens with different compositions in the future studies.

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