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Magnetic response of ultrathin Fe on MgO: A polarized neutron reflectometry study

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The magnetization of ultrathin bcc Fe films (two and three monolayers) on MgO was measured and compared with the behavior predicted for a two-dimensional ferromagnet. The experiment indicated that no hysteresis was present in the magnetization. Instead, the magnetization at low temperature was affected by a marked field cooling effect. These observations lead to the conclusion that films of Fe on MgO of such thickness exhibit superparamagnetic behavior as if they were not entirely continuous. In contrast, films thicker than five monolayers exhibit a magnetic response close to that of bulk iron.

I. INTRODUCTION

A polarized neutron reflection (PNR) study of thin films of bcc iron on MgO recently published[1] showed some surprising features. Even the thinnest films (two monolayer thick) were found to be ferromagnetic. At low temperature a sizeable magnetic field (of the order of 1 kOe) was necessary to saturate the in-plane magnetization, while fields of a few oersted were sufficient to saturate thicker samples. The amplitude of the ferromagnetic moment was found to be 2.2 ± 0.2 μ_B/Fe atom regardless of the sample thickness, in contrast with a predicted enhancement[2] for the surface atoms close to 3 μ_B/Fe atom. In view of the unusual magnetization curve at low temperature the question was raised if these thin films of iron showed the elusive magnetic behavior expected for a two-dimensional ferromagnet.

A magnet in two dimensions differs in significant ways from its three-dimensional counterpart. The Mermin–Wagner theorem shows that in the absence of anisotropy there is no magnetic ordering at zero field.[3] At finite fields the field and temperature behavior of the magnetization is governed by the equation[4]

$$\frac{M}{M_0} = 1 + \frac{k_B T}{2\pi J} \ln[1 - \exp(-2\mu H/k_b T)]. \quad (1)$$

For $\mu H < kT$ the magnetization follows a loglike behavior. Over a large temperature range the magnetization induced at a given field decreases almost linearly with increasing temperature.

The technique used was PNR, the working of which has already been discussed in detail in the literature.[5] Here it was used to measure the magnetic moment, functioning as a sensitive magnetometer. The physical quantities observed by PNR, however, differ somewhat from those observed by regular magnetometry. In PNR it is assumed that the films are formed of uniform and flat layers. If the films are not entirely uniform, the mean amplitude has to be taken for each height in the film; the *roughness* also causes some of the neutrons to be scattered out of the specular beam. Second, only the component of the magnetization in the plane of the sample is measured. This component, however, can be obtained as an *absolute* value.

II. EXPERIMENT

The samples studied consisted of the equivalent of two, three, or eight monolayers of Fe evaporated onto the substrate at room temperature. The Fe was covered by a wedge-shaped coating of gold, with a mean thickness of 200 A. These samples had been used in a previous PNR experiment,[1] and they were similar to others used in extensive magneto-optical Kerr effect measurements.[6] However fresh samples, sputtered on MgO and covered with MgO as well, showed similar magnetic behavior. The measurements were taken at temperatures ranging from 25 to 300 K and magnetic fields from 20 to 7000 Oe at the reflectometer "POSY-I" at the Intense Pulsed Neutron Source at Argonne National Laboratory. Each data point presented here has been extracted from a measurement which took approximately 12 h.

Fitting the neutron reflectivity data, we obtain a saturated moment of $2.0 \pm 0.2 \mu_B$/Fe atom, showing no enhancement over the bulk value. Demagnetizing effects do not play a role since at this field the moment lies along the applied field direction.

Figure 1 shows the temperature variation of the magnetization of the three monolayers sample at 7 kOe. The magnetization decreases linearly with increasing temperature with a slope far greater than that of bulk iron. Is this the signature of a two-dimensional magnet? The low temperature magnetization curve (Fig. 2) shows saturation at about 1000 Oe and could not be fit to a log function. On the other hand, the magnetization had features not expected for a conventional ferromagnet. No evidence for hysteresis was found, as it was checked by reversing the field and then measuring the remnant magnetization. In addition, a very marked field cooling effect was present. On cooling from 275 K in a field of 7 kOe (FC), the remanent magnetization was about half the saturation value. The remanent magnetization dropped dramatically by cooling from room temperature in zero field. The two monolayers sample displayed essentially the same features as the three monolayer sample, but with worse statistics. The eight monolayer sample showed a clear hysteresis loop, with a H_c of 50 Oe at 35 K

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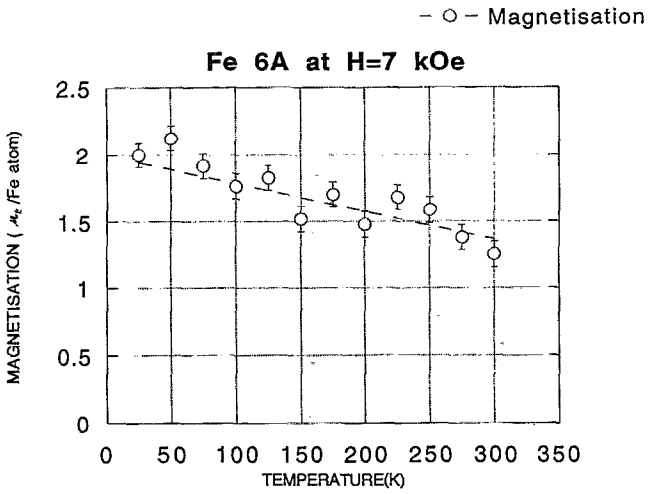

FIG. 1. Temperature dependence of the magnetization for the three monolayer sample. Data were taken at a field of 7 kOe. The dashed line is the Langevin function for particles of 1000 atoms.

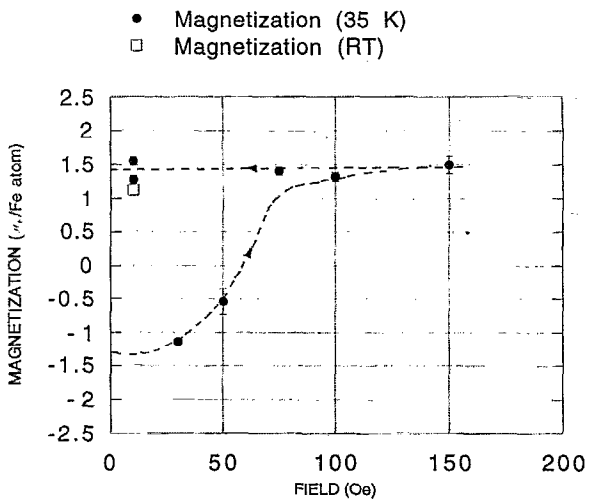

FIG. 3. Hysteresis loop at 35 K for the eight monolayer sample.

(see Fig. 3) and no appreciable variation with temperature of the magnetization at saturation.

III. DISCUSSION

The absence of hysteresis is indicative of superparamagnetism. Instead of a continuous thin film of Fe, the Fe forms islands on the surface of MgO. In a superparamagnetic material, in the absence of anisotropy, the component of magnetization in the field direction follows the Langevin function[4]

$$\frac{M}{M_0} = \coth\left(\frac{\mu H}{k_B T}\right) - \left(\frac{k_B T}{\mu H}\right), \tag{2}$$

where μ refers to the magnetization of the superparamagnetic particle, comprising a large number of atoms. Using the absolute value of the magnetization per Fe atom (obtained from fitting the neutron reflectivity data), we obtain the best fit to a Langevin function for islands of 1000 atoms in size. The calculated magnetization is presented in the form of dashed curves in Figs. 1 and 2; in Fig. 1 it is apparent that, even for superparamagnetic particles, the temperature variation of the magnetization is almost linear in a region not too close to the Curie temperature.

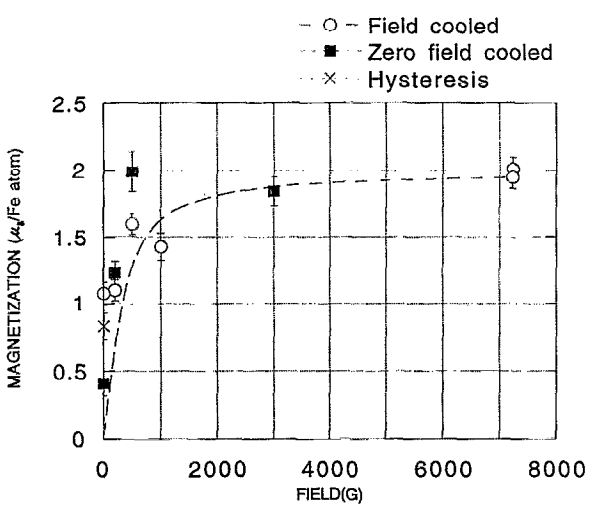

FIG. 2. Field dependence of the magnetization at 25 K of the three monolayer sample. The dashed line is the Langevin function for particles 1000 atoms in size.

According to the Langevin function the magnetization is null at zero field. However it is well known[4] that below a blocking temperature field cooling effects are present, which are interpreted as due to anisotropy. The anisotropy energy provides an energy barrier against rotation of the magnetization. If the sample is cooled in a magnetic field, and the the magnetic field is turned off, the magnetization relaxes exponentially with a time constant that is large well below the blocking temperature. A naïve calculation starting from the crystalline anisotropy of Fe gives a relaxation rate at 25 K of $\sim 10^{-9}$ s—a value which is far too small. Published measurements by Xiao *et al.*[7] on granular films confirm that the crystalline anisotropy is only a small contribution to the anisotropy energy barrier in superparamagnetic systems. We know that other anisotropies are present in our system, for instance shape anisotropy. The iron clusters are in reality thin flat islands; if they were not so, their magnetic moment would not have contributed appreciably to the magnetic reflectivity. As corroborating evidence, the magnetization of the eight monolayer Fe coverage seems to be that expected of a *continuous* film. What is not known is the detailed nature of these islands, and for that reason it is not possible at present to do further modeling: the notion itself of superparamagnetism is qualitative (because no interaction is assumed between the islands) and based on a a limited amount of observations. However, transmission electron microscopy characterization may allow us to make a more quantitative analysis.

The present study shows that films of Fe on MgO below a certain thickness do not have long range ferromagnetic ordering, such as has been observed for ultrathin Fe films on Cu,[8] Ag,[9] and Au.[10] The magnetic response can be explained in terms of superparamagnetism, which leads to the conclusion that, below a certain thickness, Fe on MgO aggregates in islands. This conclusion had been tentatively reached already by Liu *et al.*[6] on the basis of some magneto-optic measurements: below a thickness of four monolayers Fe films on MgO showed no Kerr effect signal. The lattice mismatch between MgO and Fe is only 4%; however, the difference in the surface energies (4010 nJ/mm^2 for Fe vs 1200 nJ/mm^2 for MgO) is large, which may account for the fact that at small thicknesses the Fe does not wet the MgO surface. Finally, the present measurements indicate that the magnetic moment of iron in these samples is not enhanced compared to the bulk value which is consistent with our conclusion that we have aggregates of particles in these samples.

ACKNOWLEDGMENTS

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