GRANULAR Fe METAL FILMS *

C.L. CHIEN, S.H. LIOU and Gang XIAO

Department of Physics and Astronomy, The Johns Hopkins University, Baltimore MD 21218, USA

Granular Fe metal films consisting of ultrafine Fe granules imbedded in a SiO_2 matrix have been fabricated. The granular microstructure has been revealed by transmission electron microscopy (TEM). Electron diffraction indicates that the Fe granules are bcc α-Fe and the SiO_2 matrix is amorphous. ^{57}Fe Mössbauer spectroscopy indicates that there are no measurable traces of Fe oxides. The magnetic properties of the granular Fe films deviate from those of bulk Fe due to single domain effects.

Granular metal films consist of ultrafine metal granules imbedded in an immiscible and insulating matrix [1]. The granule size can be varied from tens to thousands of Å by appropriately changing the fabrication conditions. The insulating medium insures the integrity of the ultrafine metal particles as well as separating the granules. The properties of the granular films depend sensitively on the average granular size and the volume fraction of the metal content.

We have studied granular transition metal films using a number of matrices (SiO_2, Al_2O_3, MgO, etc.). Samples were made by using dc or rf sputtering. The thicknesses of samples are about 2–5 μm. In this work, we will describe the microstructure and the magnetic properties of a Fe–SiO_2 film.

The granular nature of the samples can be best revealed by transmission electron microscopy (TEM) on suitably prepared specimens. Fig. 1 shows the TEM micrograph of Fe–SiO_2 (52 vol% Fe) which clearly displays the Fe granules. The average granular size estimated from the micrograph is about 90 Å. Also shown is the electron diffraction pattern in which the diffraction rings can be consistently indexed to the

* Work supported by ONR contract N0001485-K-0175.

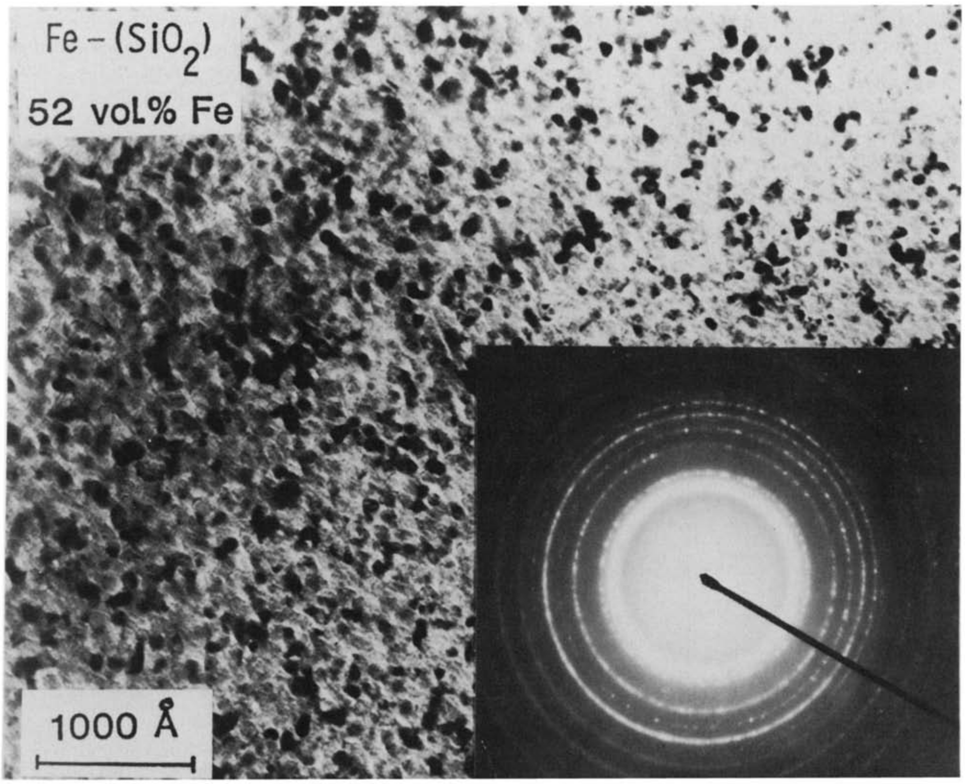

Fig. 1. TEM micrograph and electron diffraction pattern of granular Fe–SiO_2 film.

structure of bcc α-Fe. The SiO_2 medium, however, turns out to be amorphous; only its first diffraction peak, giving rise to the halo ring, is clearly observable.

Of considerable interest is the purity of the Fe granules. Electron diffraction may not detect small amount of Fe oxides. Other analytical tools which are sensitive to oxygen, such as Auger spectroscopy, will be useless since the matrix itself is oxygen rich. However, ^{57}Fe Mössbauer Spectroscopy, which measures only the Fe-containing phases and is transparent to others, is well suited for such purpose. A Mössbauer spectrum of granular Fe–SiO_2 at 4.2 K is shown in fig. 2. There is no sign of any observable amount of oxides and the effective magnetic hyperfine field of an Fe granule is the same as that of bulk α-Fe. This should be contrasted with some reports of granular Fe-containing films in which significant amounts of Fe oxides have been found [2].

Although the spectrum shown in fig. 2 is very close to that of bulk α-Fe, there are noticeable differences at 4.2 K and particularly at higher temperatures as will be detailed elsewhere.

Fig. 3a shows the magnetization at 50 kG and the remanent magnetization of the sample as a function of temperature. The measurements were made with a SQUID magnetometer. The magnetic field was applied in parallel to the sample plane. The magnetization of the sample at 50 kG which is close to the saturation magnetization of α-Fe decreases about 8% from $T = 5$ to 300 K, while the remanent magnetization decreases to near zero over the same temperature range. This is the indication of the superparamagnetism in the sample. At low temperature, thermal energy can not overcome the anisotropy (shape or crystalline) energy barriers of the single domain granules, the Fe granules then have a large magnetic remanence. As the temperature increases, thermal disturbance causes the Fe granules to exhibit superparamagnetism and decrease the remanence. Since granular size has a distribution, magnetic

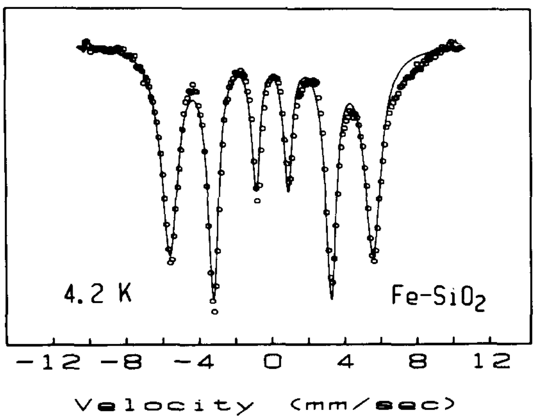

Fig. 2. Mössbauer spectrum of granular Fe–SiO_2 film at 4.2 K.

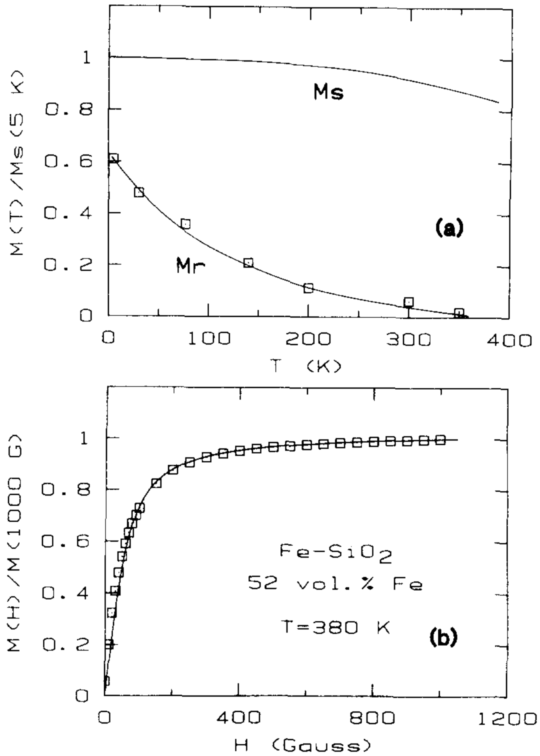

Fig. 3. (a) The magnetization at 50 kG (Ms) and the remanent magnetization (M_r) of a Fe–SiO_2 (52 vol% Fe) film as a function of temperature; (b) the normalized magnetization of the Fe–SiO_2 film as a function of external field at $T = 380$ K. The solid curve is the fit of the Langevin equation.

remanence decreases gradually as temperature increases. If all the Fe granules are superparamagnetic, the magnetization of the Fe granules can be described by the Langevin equation [3]:

$$M = vM_s\left(\coth\frac{\mu H}{kT} - \frac{kT}{\mu H}\right),$$

where v is the volume fraction of the Fe granules, μ is magnetic moment of a single granule, M_s is the saturation magnetization. Fig. 3b shows the normalized magnetization of Fe–SiO_2 (52 vol% Fe) film as a function of external field at T = 380 K which is high enough for most Fe granules to behave superparamagnetically. The fitting to the experimental data with the Langevin equation is satisfactory although a uniform granular size and spherical shape are assumed. In this fitting, the granular size was estimated to be 160 Å in diameter which is somewhat larger than that estimated from TEM micrograph.

[1] See e.g. B. Abeles, in: Applied Solid State Science, vol. 6, ed. R. Wolfe (Academic Press, New York, 1976) p. 1.

[2] J. Cui, J.L. Dormann, C. Sella and P. Renaudin, IEEE Trans. Magn. MAG-18 (1982) 1589.

[3] A. Morrish, The physical principles of magnetism (John Wiley, New York, 1965) p. 362.