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HAL Id: jpa-00219736
https://hal.archives-ouvertes.fr/jpa-00219736
Submitted on 1 Jan 1980

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MÖSSBAUER SPECTROSCOPIC STUDIES OF MAGNETIC PROPERTIES OF ULTRA-THIN FERROMAGNETIC FILMS

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For several years we have been investigating the magnetic properties of epitaxial [110] iron films in the thickness range from 20 Å to 100 Å. These films were grown on [111] Ag, in ultra-high vacuum, and were investigated for purity by layer-by-layer argon sputtering combined by Auger spectroscopy.

A typical Mössbauer spectrum for a thin sample is shown in Fig. 1. Measurements of magnetic hyperfine field at several temperatures from 4.2 K to about 500 K are shown in Fig. 2. Several features are apparent:

First, the hyperfine field (magnetization) decreases more rapidly at higher temperatures for the thinner films. This is predicted by all models of collective magnetic excitations. Second, the variation of magnetization with temperature from 4.2 K to 300 K is approximately linear for the 20 and 30 Å films, while the thicker ones show the more usual $T^{3/2}$ variation. The linear behavior has been predicted by several investigators, the most recent of whom were Levy and Motchane. Their result is actually $\Delta M(T) \propto T \text{ e}^{-E_a/kT}$, where $E_a$ is an anisotropy energy. For $E_a \ll kT$ the behavior is approximately linear, as observed.

The third interesting feature is the apparent increase in magnetic hyperfine field at 4.2 K for the thinner films. The precise explanation of this phenomenon was not immediately apparent, but an additional piece of evidence was the isomer shifts of the Mössbauer hyperfine spectra. In our spectra, at all temperatures, we observed that the electron density, as measured in this way, was less for the thinner films that for thicker ones. We concluded that both the increased hyperfine field at low temperatures and the decreased electron density at the nucleus have the same cause: namely, a different electronic structure for magnetic atoms near the surface of the films. We can now explain all of the unusual features of the hyperfine field (magnetization) curves shown in Fig. 2. The more rapid decrease of magnetization with temperature for the thinner films results from the smaller number of exchange interactions for atoms near the surface of

* Work supported by NSF Grant DMR77-07127-AO1
the films. This size, or thickness, effect should disappear at T=0. Finally, the surface atoms and their nearest neighbors have a different electronic structure near the surface. The conduction electron density is lower there and the magnetic hyperfine field, and perhaps the magnetic moment per Fe atom, are affected so that at T=0 a larger hyperfine field than that of bulk Fe is observed.

While this is a consistent qualitative picture we were concerned to have additional evidence. To obtain this information we used the fact that $^{56}\text{Fe}$, although magnetically identical to the Mössbauer isotope $^{57}\text{Fe}$, shows no Mössbauer effect itself. We obtained isotopically pure $^{56}\text{Fe}$ and constructed a series of epitaxial films all of nominal thicknesses of about 60 Å. In each film a probe layer of 4-10 Å thickness of $^{57}\text{Fe}$ was deposited, with the different films having the probe layer at different depths in the film. In this way we could probe the magnetic hyperfine fields and electron densities at different places in the film as a function of temperature. The interesting results are shown in Fig. 3, showing data taken at 295 K and at 4.2 K.

Fig. 3 Hyperfine field vs. $^{57}\text{Fe}$ probe position in a family of 30 layer Fe films (at 295 and 4.2 K). In each case we show the magnetic hyperfine field (in kilogauss) measured for each probe layer, as a function of the position of the probe layer in the nominally 30 layer films.

Despite the obvious experimental difficulties it can be seen that the earlier conjecture (that the unusual magnetic behavior was concentrated at the surface of the film) is valid. The electron density determinations show a fall-off in density near the surface qualitatively similar to that of the hyperfine field at 295 K. The increase in hyperfine field near the surface at 4.2 K is also clearly seen.

What remains to be understood is whether these surface effects are primarily the result of the existence of a surface of magnetic atoms, or the result of electronic interaction between the surface iron atoms and the silver covering layer used to prevent oxidation.

We have also determined the variation of critical temperature, $T_C$, with film thickness. Fisher$^2$ and others$^3$ have predicted a relation $[T_C(\infty) - T_C(L)]/T_C(\infty) = C/L^\lambda$ where $L$ is the thickness of the film. The constant $C$ depends on the scale of the exchange interaction in terms of $L$. The critical exponent $\lambda$ is expected to be $1/\nu$ for short-range models (Ising model) and 1 for long-range models (mean field theories). Here $\nu$ is the critical exponent which determines the correlation length near $T_C$. It is known to be about 2/3. From our measurements determining $T_C$ by the temperature at which all hyperfine splitting collapses, we obtain a value of $\lambda = 1.6 \pm 0.1$, consistent with a short-range scaling theory.

References