Magnetism of Epitaxial bcc Iron on Ag(001) Observed by Spin-Polarized Photoemission

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Epitaxial bcc Fe films grown on Ag(001) are ferromagnetic. At $T=30$ K, the 3- to 4-monolayer films are perpendicularly magnetized at remanence, whereas thinner and thicker films have their magnetization in plane. Above $T=100$ K no perpendicular remanence has been observed for any film thickness. Films consisting of more than 5 monolayers have a Curie temperature equal to that of bulk bcc Fe.

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The feasibility of growing perfectly epitaxial metallic overlayers has opened the way for studying new phases of materials which under normal conditions do not exist as three-dimensional solids. Examples are fcc cobalt and fcc iron, which can both be grown on Cu(001).[1,2] Beside the interest in the study of unusual forms of matter, there is another equally fascinating aspect of thin epitaxial films: the occurrence of specific two-dimensional features not encountered in bulk specimens. Recently, much debate arose on the magnetization properties of thin epitaxial bcc Fe films on Ag(001) [abbreviated bcc Fe/Ag(001)].[3,4] This system forms the subject of this paper.

The surface sensitivity of the spin-polarized photoemission experiment[5] was exploited to study the magnetization of epitaxial bcc Fe/Ag(001) films in an external field at various temperatures. The field was applied perpendicular to the plane of the films. The film thickness varied between 0.8 and 10 monolayers (ML). In contrast to fcc Fe on Cu(001),[2] no upper limit for the thickness of epitaxial bcc Fe overlayers on Ag(001) has been found. This is a plausible result since the stable phase of ferromagnetic Fe is body-centered cubic.

The spin polarization of the photoemitted electrons as function of the external field, $P(H)$, is shown in Fig. 1 for bcc Fe films at $T=30$ K. There is a most striking dependence of $P(H)$ on the number of monolayers, a feature not unfamiliar from the fcc Fe/Cu(001) system.[2] Very thin films show no remanence along the surface normal. For an intermediate range of thickness—from 3 to 4 ML—there is a perpendicular remanence amounting to almost the full saturation polarization for 3.5 ML. For thicker samples, ≥ 5 ML, the remanence again vanishes. Above 100 K, no perpendicular remanence is found for all film thicknesses.

The (001) LEED patterns of fcc and bcc Fe are identical; therefore it cannot be used to distinguish between the two phases. However, the distinction is easily made with use of the temperature dependence of the spin polarization $P(T)$: For bcc Fe/Ag(001) films thicker than 5 ML, the Curie temperature T_C becomes 1000 ± 100 K (see Fig. 2), whereas T_C of fcc Fe/Cu(001) never exceeds 500 K over the thickness range which is stable (≤ 15 ML). Very thin magnetic films—in the case of Fe/Ag(001) <5 ML—have a reduced transition temperature, a fact which has been known for a long time.[6]

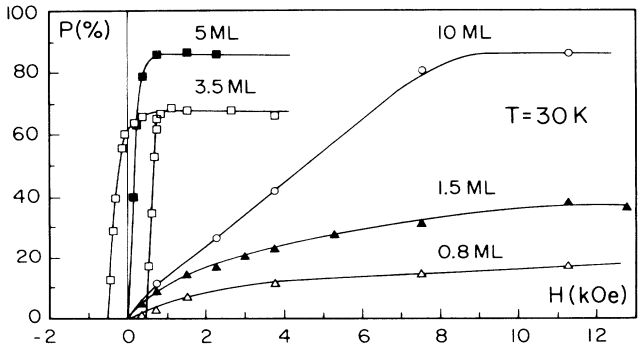

FIG. 1. Magnetization curves taken at $T=30$ K for epitaxial films of bcc Fe on Ag(001) of various thicknesses, indicated in monolayers (ML). The field H is applied perpendicularly to the surface.

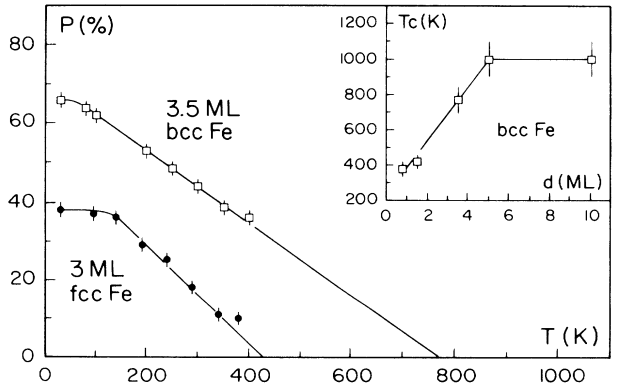

FIG. 2. Temperature dependence of the saturation polarization of a 3.5-ML-thick epitaxial bcc Fe film on Ag(001) and a 3-ML fcc Fe film on Cu(001). Inset: Thickness dependence of the Curie temperature of the bcc Fe films.

T_C of a 1-ML film is about 400 K.

The films were prepared by evaporation from a resistively heated Fe wire onto a Ag(001) crystal, the growth rate being typically 0.2 ML/min. The substrate was at room temperature. With the atoms of the first Fe layer occupying the bridge positions between the Ag atoms of the outermost layer of the substrate, the lattice spacing of the epitaxial Fe film is very nearly that of bulk bcc iron (2.89 Å vs 2.87 Å at room temperature). Film growth was monitored by use of Auger spectroscopy and LEED. The quadratic LEED pattern observed at 55 eV persisted for all film thicknesses. However, between 2 and 4 ML, the spots were slightly broader than for either thinner or thicker films. This phenomenon has been observed also in other laboratories[7] and its cause is currently being investigated. Only films >4 ML showed a slight O contamination amounting to less than 5 at.%. The absolute film thickness was determined with an estimated accuracy of $\pm 20\%$ from the ratio of the Ag 305-eV and Fe 650-eV lines. The relative determination of the film thicknesses with respect to each other is more accurate. It is well established that Fe on Ag(001) grows in a layer-by-layer mode.[3,4,8]

The full spectrum of a Hg-Xe lamp, $h\nu < 5.5$ eV, was used as light source in the photoemission experiment. The photothreshold of the films was 4.5 ± 0.05 eV. The measured quantity is the spin polarization of the emitted photoelectrons $P=(N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$, where $N_\uparrow$ ($N_\downarrow$) is the number of electrons with spin magnetic moment parallel (antiparallel) to the magnetization along the surface normal. Details on the experimental technique can be found in Ref. 5. The sample temperature was varied between 30 and 450 K. Even at 450 K, no indication of interdiffusion of Fe and Ag has been observed within the measuring time as judged from the reversibility of the $P(T)$ behavior.

From Figs. 1 and 2, the following facts are evident: (1) Films of 1 ML or thicker of bcc Fe/Ag(001) are ferromagnetic. (2) At $T=30$ K, the remanence magnetization of the 3.5-ML film is directed along the surface normal. The remanence equals practically the full saturation magnetization showing the film to be essentially single domain. (3) The Curie temperature of films thicker than 5 ML equals that of bulk bcc Fe.

For thin bcc Fe/Ag(001) films, both experiment[3,4] and theory[9] have given indications for perpendicular magnetocrystalline anisotropy tending to keep the magnetization along the surface normal. The experimental situation was, however, by no means clear since the crucial test—the measurement of the remanent magnetization along the surface normal—could not be performed because of inherent constraints of the experimental arrangements. The measurements reported in this paper are the first ones which were made by the direct observation of the component of the magnetization perpendicular to the sample surface. The result is that the perpendicular anisotropy is indeed present and under certain conditions strong enough to keep the remanence magnetization perpendicular. However, the actual behavior shows a very complex dependence of the anisotropy on film thickness and temperature.

At the measuring temperature $T=30$ K, it is found that the perpendicular remanence of very thin (<2 ML) films vanishes, as it does for films thicker than 5 ML. Two possibilities may account for this observation: Either regions of perpendicular magnetization compensate each other (as, for example, in bubble films with bias field removed) or the shape anisotropy forces the magnetization to lie in plane. For the epitaxial films studied in this paper, there are simple arguments to exclude the first possibility.

For an ideal, defect-free, perpendicularly magnetized film, the incentive to break up into domains vanishes for sufficiently small thickness d, namely when the energy gain by flux closure at a domain boundary (proportional to d^2) is overcompensated by the energy cost of the domain wall ($\simeq d$).[10,11] Any perpendicularly magnetized film consisting of a few monolayers only must be single domain. Therefore, the films in Fig. 1 with zero remanence are magnetized in plane.

A further proof of the in-plane magnetization at $H=0$ is the shape of the $P(H)$ curves; see Fig. 1. A film with perpendicularly magnetized domains shows a characteristic $P(H)$ behavior due to the peculiar development of the widths of the "up" and "down" domains in an external field. At low fields the width of the "up" domains (parallel to H) grows proportional to H; at higher fields the width increases much more rapidly, immediately before collapse of the stripes with an infinite slope. This leads to an upward-bent concave form of $M(H)$ as observed in bubble films.[11] Nothing of this kind, however, is found in case of the few-monolayer Fe/Ag(001) films, proving again that for those films of Fig. 1 without remanence the magnetization lies in plane.

A nonideal film possesses a magnetization curve influenced by irreversible effects which are characterized by a frictionlike dynamic and static wall-motion coercivity.[11] It prevents the formation of an equilibrium domian configuration, but it is evidently too weak to give rise to a measurable perpendicular remanence for all films in Fig. 1, except possibly the one with 3.5 ML. However, since the perpendicular remanence occurs reproducibly for (3.5 ± 1)-ML films, it is attributed to an intrinsic film property—uniaxial anisotropy—and not to the spurious occurrence of defects.

It should be noted that it may be questionable to extrapolate concepts used for bubble films[10,11] to the interpretation of measurements on films as thin as those studied in this paper. Then, a better understanding must be based on a direct quantum-mechanical treatment[9] or use of a suitable model calculation.[12]

It is an intriguing problem why only for (3.5 ± 1)-ML bcc Fe/Ag(001) films is the perpendicular anisotropy strong enough to overcome the demagnetizing energy (or shape anisotropy) which favors in-plane magnetization. A likely cause may be found in the microstructure of the Fe/Ag interface. Magnetic anisotropies are very sensitive to the nearby atomic environment (stress and strain). Therefore a very precise knowledge of the atomic positions and binding properties within the interface seems indispensable for getting insight into the peculiar behavior of these anisotropies.

The temperature dependence $P(T)$ is shown in Fig. 2 for both a 3-ML fcc Fe/Cu(001) film and a 3.5-ML bcc Fe/Ag(001) film. The extrapolated Curie temperatures differ by almost a factor 2. The 5-ML bcc Fe/Ag(001) film already possesses the Curie temperature of bulk iron. By measurement of T_C, bcc and fcc Fe films are easily distinguished—in contrast to LEED studies where the two structures give identical patterns though with different $I(V)$ characteristics. The structural instability of thick (>15 ML) fcc Fe/Cu(001) was detected by a jump of T_C to about 1000 K after a spontaneous transformation into the stable bcc phase.[13] The linear dependence of P on T for surfaces of bulk solids and the dependence of T_C on film thickness are well-known phenomena which have been observed for many different systems.[6] However, little is known about $P(T)$ of films only a few monolayers thick. For Fe on Cu(001) (Ref. 2) and for Fe on Ag(001), $P(T)$ also appears to be linear. Since the polarization is measured in an external field, T_C is extrapolated from the $P(T)$ data with T sufficiently far away from T_C where the magnetization is determined by the exchange field alone. Note that the same procedure is used for the determination of T_C of bulk specimens.[14]

Above 100 K, no hysteresis behavior of $P(H)$ has been found for all film thicknesses.[13] For the same reasons as given above for the $T=30$-K measurements, M is thought to lie in plane at $T>100$ K.

Another remarkable fact which deserves future investigation is the high threshold polarization of 85% observed in films which are thicker than about 5 ML. This is substantially more than the 60%–65% measured with the same equipment on Fe(111)[15] or polycrystalline Fe.[16] Since the measurement is energy and angle integrated with light energies up to 1 eV above photothreshold, P reasonably represents a density-of-states polarization independent of the particular surface studied.

Epitaxial magnetic films present a largely unexplored but, nowadays, experimentally accessible field of magnetism. Although the technique of producing metallic interfaces and their characterization is still in its infancy, it offers the possibility of the creation of clean and highly reproducible magnetic structures. This may also be of importance for application-oriented research, as, e.g., thermomagnetic recording which has been proved feasible on 5-ML Fe/Cu(001) films.[17] The infinitely varied world of magnetism offers yet another exciting prospect for the future.

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