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Magnetic ordering and anisotropy of epitaxially grown Fe_xCu_{1-x} alloy on GaAs(001)

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Single crystalline Fe_xCu_{1-x} alloys over the entire composition range have been prepared successfully on GaAs(001) via molecular beam epitaxy. The films are body centered cubic (bcc) at high Fe concentration ($x > 0.75$), body centered tetragonal (bct) or face centered cubic (fcc) at the lower Fe concentration depending on the film thickness. Long-range ferromagnetic order is observed when $x > 0.33$ where the thickness is 6 nm, and the effective magnetic moment per Fe atom decreases as the Fe concentration increases. Fe-Cu alloy films with four-fold magnetocrystalline anisotropy were obtained.

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The alloying effect on the structure and magnetism of transition metals has long been an interesting topic. In particular, much attention has been paid to the Fe_xCu_{1-x} alloy[1–4] because of the tremendous interest in the magnetism of face-centered-cubic (fcc) Fe,[5] for it can only be prepared via precipitation in a Cu matrix or epitaxially grown on a Cu substrate. On the other hand, because of the extreme low bulk miscibility of either Cu in Fe or Fe in Cu, only ~4% Fe dissolves into Cu (with fcc structure) and ~10% Cu into Fe (with bcc structure) near their respective liquid points,[6] a long-standing challenge has been the goal of producing metastable solid solutions with higher concentrations by different approaches, such as rapid quenching, vapor deposition, ion beam mixing, or mechanical alloying.[7–11]

With regard to the structure, it is known that Fe_xCu_{1-x} alloy can be obtained by: (i) the rapid quenching method with fcc structure at $x < 0.20$, bcc structure at $x > 0.85$, and mixed fcc and bcc phase at $0.20 < x < 0.85$;[12] (ii) the vapor deposition method with fcc structure at $x < 0.40$, bcc structure at $x > 0.60$, and mixed fcc and bcc phase at $0.40 < x < 0.60$,[7] or with fcc at $x < 0.60$, bcc at $x > 0.75$, and mixed fcc and bcc at $0.60 < x < 0.75$;[8] (iii) the mechanical alloying method with fcc structure at $x < 0.60$, bcc structure at $x > 0.70$, and mixed fcc and bcc phase at $0.60 < x < 0.70$,[13] or with fcc at $x < 0.60$, bcc at $x > 0.80$, and mixed fcc and bcc at $0.60 < x < 0.80$.[14] However, all the samples prepared in these works are polycrystalline. The only exception reported so far is a monolayer stacked Fe/Cu superlattice fabricated on a Cu(100) substrate by laser pulsed deposition, yet corresponding to only one particular composition at $x = 0.5$, $Fe_{0.5}Cu_{0.5}$.[15] A natural though quite ambitious question to be asked is whether there is a way to get the single crystalline Fe_xCu_{1-x} alloy over the entire composition range.

On the magnetism, it is clear that the magnetic ordering of Fe_xCu_{1-x} alloy changes from nonmagnetic to ferromagnetic states as the Fe content x increases. However, how the magnetic moment of Fe changes as a function of the compositions is rather controversial. Experimentally, Uenishi *et al.* showed that the Fe moment remains $\sim 2.2\mu_B$/atom over a wide composition range at $x > 0.50$, then quickly falls to zero as x decreases,[13] but Chien *et al.* reported that there were no noticeable changes in the magnetic moment over the entire composition range.[8] Theoretically, on the other hand, Serena *et al.* showed with the first principles calculation that the magnetic moment per Fe atom was almost independent of Fe-richness for a wide range of Fe content ($x > 0.5$) in both bcc and fcc phases, then decreased and reached to zero for $x \leqslant 0.25$.[16] However, Wang *et al.* reported, after taking into account the magneto-volume effect, that the magnetic moment per Fe atom in the bcc phase increased from 2.2 μ_B to 2.62 μ_B as the Fe concentration x decreased from 1 to 0.5.[17] It is thus interesting to see what would happen once better defined single crystalline Fe_xCu_{1-x} films are obtained. In addition, another issue which is important but has never been addressed is whether the magnetocrystalline anisotropy exists in the single crystalline, but chemically disordered, Fe_xCu_{1-x} films.

In this work, we demonstrate that single crystalline Fe_xCu_{1-x} alloy over the entire x range can indeed be grown epitaxially on a GaAs(001) substrate. The structure of Fe_xCu_{1-x} is shown to be a function of film thickness as well as Fe composition. It is found that the magnetic moment per Fe atom in the films becomes larger as the Fe concentration decreases from 1 to 0.5. A fourfold magnetocrystalline anisotropy is observed in the Fe_xCu_{1-x} alloys at $x \geqslant 0.5$, together with a uniaxial magnetic anisotropy, which is generally believed to be caused by the GaAs(001) substrate.

The detailed description about the experimental setup and procedure can be found in our previous works,[18,19] so it will not be repeated here. 99.99% pure Fe and Cu metal pieces were used as the evaporating sources. The Fe_xCu_{1-x} films were prepared by co-evaporation on the GaAs(001) substrate at room temperature. The deposition rate of Fe and Cu were calibrated separately by a quartz microbalance, and the overall co-evaporating rate was controlled at about 0.2 nm/min.

It is well known that Fe can be epitaxially grown on GaAs(001) with the bcc structure.[20] Here we give in Fig. 1(a) a typical reflection high energy electron diffraction (RHEED) pattern for the bcc-Fe, with the electron incidence along the [110] direction of GaAs(001). A detailed description of how to distinguish the fcc, bct, and bcc structures

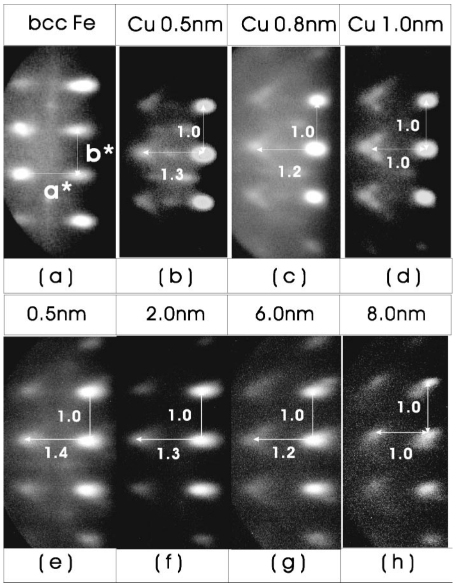

FIG. 1. RHEED patterns for (a) Fe film, (b)–(d) Cu film, and (e)–(h) $Fe_{0.25}Cu_{0.75}$ film on GaAs(001).

from a RHEED pattern can be found elsewhere.[18] It can be seen that the aspect ratio of a^*/b^* in the reciprocal space is close to 1.4, as it should be expected. On the other hand, since the lattice mismatch between bcc-Cu and GaAs(001) (1.2%) is significantly smaller than that between fcc-Cu and GaAs(001) (9.7%), an epitaxial growth of bcc-like Cu on GaAs(001) would presumably be realized in experiment. In fact, we have realized it on a GaAs(001) substrate for the first several layers, as shown in Fig. 1(b). As the film thickness increases, it is found that the diffraction pattern becomes more and more square-like, i.e., the aspect ratio of a^*/b^* gradually decreases and finally reaches to 1, as shown in Figs. 1(c) and 1(d). This result indicates that Cu on GaAs(001) is in an almost bcc like structure at the beginning, then in body-centered-tetragonal (bct), and finally in its thermodynamically stable fcc structure. Based on the foregoing results at the two extremes of $x=1$(Fe) and $x=0$ (Cu), it seems very promising to obtain the single crystalline epitaxial films of Fe_xCu_{1-x} over the entire composition range $0<x<1$.

For the growth of Fe_xCu_{1-x} on GaAs(001) with any particular x value, similar to the case of Cu on GaAs(001), it is noted that the structure varies with the film thickness. As a representative example, we show in Figs. 1(e)–1(h) the structural evolution as a function of film thickness for the growth of $Fe_{0.25}Cu_{0.75}$. It has a bcc structure at the initial stage of growth, then changes to a bct (closer to bcc) structure, then to a bct but closer to fcc structure, and finally to a fcc structure. For a summary of the growth of Fe_xCu_{1-x} on GaAs(001) at different x values we show in Fig. 2 the structure (a^*/b^*) versus film thickness at different Fe composition x. It is seen that at the same thickness of 6 nm, the (a^*/b^*) ratio is 1.4 (bcc) at $x=1$,1.3 at $x=0.75$,1.25 at $x=0.5$,1.2 at $x=0.25$, and finally 1.0 (fcc) at $x=0$(Cu). This tendency is actually quite consistent with the fact that bcc is the stable phase for Fe, while fcc is the stable phase for Cu, so the structure of Fe_xCu_{1-x} at any x should be determined by the competition between these two factors.

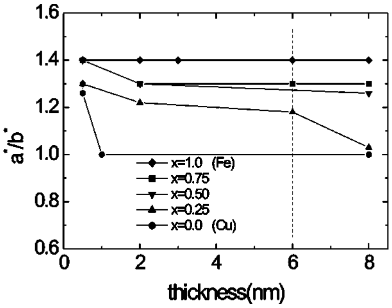

FIG. 2. The a^*/b^* aspect ratio from RHEED pattern changes as a function of the film thickness for Fe_xCu_{1-x} at different x.

The samples prepared in ultra high vacuuam were covered by 2 nm gold before being taken out for the magnetic measurements. Using the surface magneto-optical Kerr effect (SMOKE) technique measured for 6-nm-thick films at room temperature, we find that an Fe-rich Fe_xCu_{1-x} alloy $(x>0.35)$ is ferromagnetic while an Fe-poor Fe_xCu_{1-x} alloy $(x<0.30)$ is nonmagnetic, and the transition happens around $x=0.33$. This result is quite different from that found previously in comparable polycrystalline samples, where the transition was seen around $x=0.5$.[16] This is presumably caused by the different structures realized in the experiments, yet a clear explanation still needs to be worked out.

As mentioned previously, the film structure of Fe_xCu_{1-x} alloy on GaAs(001) depends on the film thickness, so it is important to check the effect of thickness on the magnetization. Figure 3(a) gives the result for Fe_xCu_{1-x} alloy at $x=0.5$, measured in a wedged sample at room temperature. It is seen that there is no noticeable change of effective magnetic moment per Fe atom at room temperature, otherwise the Kerr intensity versus thickness would not be such a nice straight line. On the other hand, the finite size effect on the Curie temperature does exist in the ultrathin regime below 2.0 nm,[21] where the deviation from the straight line is clearly seen. Since SMOKE does not provide quantitative information about magnetization, a superconductor quantum interfer-

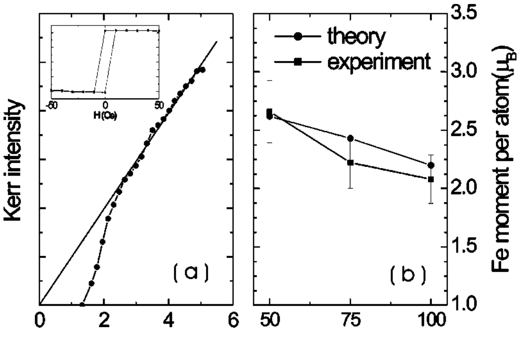

FIG. 3. (a) The Kerr intensity at saturation is plotted as a function of film thickness for $Fe_{0.5}Cu_{0.5}$. The inset gives a typical MOKE hysteresis loap, from which the Kerr intensity at saturation is obtained; (d) the effective magnetic moment per Fe concentration.

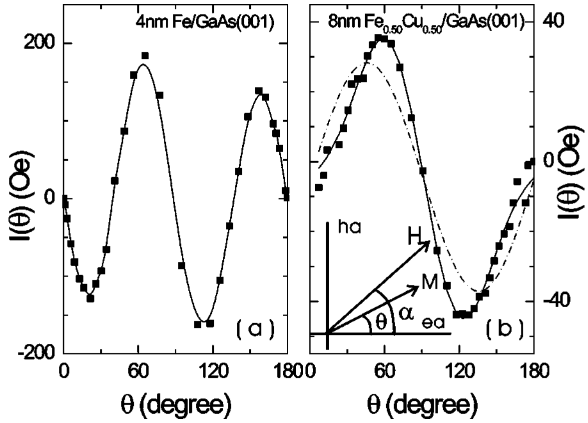

FIG. 4. The torque moment $l(\theta)$ as a function of θ, (a) $H=800$ Oe, 4 nm Fe; (b) $H=200$ Oe, 8 nm Fe$_{0.50}$Cu$_{0.50}$. A schematic picture of the applied field H and the magnetization M is shown in the inset.

ence device (SQUID) measurement is carried out at 5 K to determine the effective magnetic moment per Fe atom for 6-nm-thick Fe$_x$Cu$_{1-x}$ films, assuming that the contribution of Cu to the total magnetization can be neglected. The results are summarized in Fig. 3(b). It is seen that the effective magnetic moment per Fe atom decreases when the Fe$_x$Cu$_{1-x}$ alloy becomes more and more Fe-rich, a result consistent with that predicted earlier by Wang *et al.*,[17] which is also shown in the figure. It should be mentioned that the total magnetization is too small to be detected in the SQUID measurement for the Fe$_{0.33}$Cu$_{0.67}$ film when the thickness is 6 nm, therefore the moment per Fe atom is not shown.

The success in producing single crystalline Fe$_x$Cu$_{1-x}$ alloy on GaAs(001) makes the magnetic anisotropy measurement possible. Although Fe on GaAs(001) exhibits a well-defined fourfold in-plane magnetic anisotropy, together with a uniaxial anisotropy believed to be induced by the substrate,[22] it is not trivial to consider whether similar magnetic anisotropy would also exist in Fe$_x$Cu$_{1-x}$ on GaAs(001). To address this problem, longitudinal MOKE in combination with a rotating magnetic field, called the ROTMOKE method,[23] is used here to determine the magnetic anisotropy of the Fe$_x$Cu$_{1-x}$ films. We will follow the treatment given by Mattheis *et al.*,[23] except that we have to consider not only the uniaxial but also the fourfold anisotropy.

Based on the fact that Fe grown on GaAs(001) has a fourfold anisotropy with the easy axis along the $\langle 100\rangle$ direction and a uniaxial anisotropy with the easy axis along the $[1\bar{1}0]$ direction,[24] the total energy density of the system is

$$E/V = -M_sH\cos(\alpha-\theta) + K_1\sin^2\theta + K_2\sin^2(\theta+45°)\cos^2(\theta+45°). \tag{1}$$

Here, the first term represents the Zeeman energy under the applied magnetic field H, M_s is the saturation magnetization, α is the angle between the easiest axis (ea) and the applied magnetic field [see the inset of Fig. 4(b)], θ is the angle between the easiest axis and the magnetization; the second term is the uniaxial anisotropy energy, and the third term is the fourfold anisotropy energy; V is the volume of the magnetic film. According to Mattheis *et al.*,[23] a Stoner-Wohlfarth like magnetization reversal process can be realized at sufficient large magnetic field. Neglecting any hysteresis one can expect that the magnetization takes the equilibrium angle θ, which can be obtained by differentiating Eq. (1):

$$HM_s\sin(\alpha-\theta) = K_1\sin 2\theta - (K_2/2)\sin 4\theta. \tag{2}$$

Keeping in mind that the term

$$L[\alpha(\theta)] = HVM_s\sin(\alpha-\theta)$$

is a torque, which is usually determined by torquemetry,[25] one is able to determine in experiment a related value $\{l[\alpha(\theta)]=L[\alpha(\theta)]/VM_s\}$ from ROTMOKE by simultaneous determination of the two angles α and θ. Then by plotting $l(\theta)$ as a function of θ, one can estimate experimentally the anisotropy of the system under consideration. Furthermore, after fitting the $l(\theta)$ curve with the following [from Eq. (2)]:

$$l(\theta) = H\sin(\alpha-\theta) = (1/2)H_{k_1}\sin 2\theta - (1/4)H_{k_2}\sin 4\theta, \tag{3}$$

one can finally obtain the anisotropic fields $H_{k1}=2K_1/M_s$ and $H_{k2}=2K_2/M_s$.

By fitting the curve using Eq. (3) (solid line), the uniaxial and fourfold anisotropic fields have been determined to be $H_{k1}=62$ Oe, $H_{k2}=604$ Oe, respectively, for 4 nm pure Fe/GaAs(001). Figure 4(a) shows the measured torque moment $l(\theta)$ (by dots) as a function of angle θ, at $H=800$ Oe, for pure Fe on the GaAs(001) sample. On the one hand, a periodicity of 90° in the oscillation curve is clearly seen, which indicates that a fourfold magnetic anisotropy does exist. On the other hand, it is noted that the values of $l(60°)$ and $l(150°)$ are not the same, indicating that a uniaxial magnetic anisotropy also exists.

An Fe -rich Fe$_x$Cu$_{1-x}$ alloy can be regarded as putting Cu gradually into the Fe matrix; it is reasonable to assume the fourfold magnetic anisotropy, if existing, still remains on the same easy axis as that of Fe, thus we start with the same total energy density [Eq. (2)] to describe Fe$_x$Cu$_{1-x}$. Figure 4(b) gives the torque moment $l(\theta)$ as a function of angle θ, at $H=200$ Oe, for Fe$_{0.50}$Cu$_{0.50}$ on GaAs(001). At first glance it seems like a typical uniaxial magnetic anisotropy, because of the 180° periodicity in the oscillation. However, once the data are really fitted with the uniaxial anisotropy alone, i.e., only the first term in Eq. (3), it is realized immediately that the fitting is rather poor, as shown by the dashed line in the figure. Interestingly, the situation is significantly improved when the data are fitted with both uniaxial and fourfold anisotropies, as shown by the solid line in the figure. The uniaxial and fourfold magnetic anisotropic fields are determined to be $H_{k1}=69$ Oe, $H_{k2}=47$ Oe for 8 nm Fe$_{0.50}$Cu$_{0.50}$/GaAs(001). It is important to note that the sign of H_{k2} is positive rather than negative, indicating the same fourfold magnetic anisotropy as that of Fe. Obviously the fourfold anisotropy of Fe$_{0.50}$Cu$_{0.50}$/GaAs(001) becomes much weaker compared to that of pure Fe. However, if the content of Fe in the alloy increases, a larger H_{k2} would be expected. This turns out to be true, since $H_{k2}=172$ Oe is found for 8 nm $\mathrm{Fe_{0.75}Cu_{0.25}/GaAs(001)}$.

Based on these results we declare for that $\mathrm{Fe}_x\mathrm{Cu}_{1-x}$ alloys retain a fourfold magnetoanisotropy on GaAs(001) although it is chemically disordered, which might indicate that the magnetocrystalline anisotropy in $\mathrm{Fe}_x\mathrm{Cu}_{1-x}$ is dominated by the local spin-orbit coupling. On the other hand, the existence of the fourfold magnetic anisotropy implies the high quality of the single crystalline $\mathrm{Fe}_x\mathrm{Cu}_{1-x}$ film prepared in this work.

In conclusion, it is shown that single crystalline $\mathrm{Fe}_x\mathrm{Cu}_{1-x}$ alloy over the entire composition range can be obtained on GaAs(001). It shows ferromagnetism as $x \geqslant 0.33$ when film thickness is 6 nm and the magnetic moment per Fe atom increases as the Fe concentration decreases. A fourfold magnetocrystalline anisotropy is also realized in the $\mathrm{Fe}_x\mathrm{Cu}_{1-x}$ alloys.

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