Interface Magnetization Reversal and Anisotropy in Fe/AlGaAs(001)

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The reversal process of the Fe *interface layer* magnetization in Fe/AlGaAs heterostructures is measured directly using magnetization-induced second-harmonic generation, and is compared with the reversal of the bulk magnetization as obtained from magneto-optic Kerr effect. The switching characteristics are distinctly different due to interface-derived anisotropy—single step switching occurs at the interface layer, while two-jump switching occurs in the bulk Fe for the magnetic field orientations employed. The angle between the interface and bulk magnetization may be as large as 40–85 degrees. Such interface switching will dominate the behavior of nanoscale structures.

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The interest in single-crystal ferromagnetic thin films grown on semiconductor substrates has increased in recent years because of their potential use in next-generation spintronic devices which utilize spin injection or detection through a ferromagnetic thin-film semiconductor interface [1–3]. A single-crystal Fe(001) film on GaAs(001) is a promising spin injection heterostructure [4–7], and a large number of studies have addressed this system in recent years [8–17]. High quality epitaxial growth of Fe on GaAs(001) has been achieved due to a relatively small lattice mismatch ($2a_{\mathrm{Fe}}/a_{\mathrm{GaAs}} = 1.013$) [8,9].

Very thin Fe(001) films exhibit an unusual in-plane uniaxial component to the magnetic anisotropy, such that the easy axis is along [110] for films <15 monolayers ($\sim$21 Å) thick [11]. This is dramatically different from the cubic magnetic anisotropy of bulk bcc Fe which has $\langle 100 \rangle$ easy axes. A detailed study of Fe films grown on well-characterized GaAs(001) surfaces prepared by molecular beam epitaxy (MBE) attributed this behavior to an interface contribution derived from the directional character of Fe-As bonding [11], corroborating the original hypothesis of Krebs *et al.* [9]. It was argued that this Fe-As bonding is common to the interface which ultimately forms, regardless of the initial GaAs(001) surface reconstruction [12]. Further evidence for this picture was provided by x-ray absorption studies, which showed that Fe 3*d* charge transfer at the interface was independent of GaAs substrate preparation and orientation [18].

Such a uniaxial magnetic anisotropy (UMA) component profoundly affects the magnetization reversal process, leading to ''one-jump'' or ''two-jump'' switching depending upon the orientation of the applied field and the hard in-plane axis resulting from the UMA [19,20]. More recent work [21] showed that the shear strain introduced from anisotropic relaxation of the Fe lattice for films thicker than 20 Å produced a strain anisotropy which competes with the interface-derived contribution, significantly modifying the overall behavior.

A fundamental understanding of the evolving magnetic anisotropy remains elusive and is a critical technological issue, since the anisotropy determines the switching characteristics which control the performance of magnetic media, random access memory, and the dynamic response at the nanoscale as illustrated by recent work on spin momentum transfer [22]. Interface-derived components will, in fact, dominate the behavior of nanoscale structures. The magnetic response measured by most experimental techniques represents the average magnetization of the entire film thickness. A tacit assumption made in each of the above studies and in the standard models of thin-film magnetic anisotropy is that the magnetization of the interface is rigidly coupled to the ''bulk'' magnetization by the strong exchange coupling typical of magnetic transition metals [23]. In this Letter, we measure the Fe/AlGaAs interface magnetization directly, and provide clear evidence that the reversal process of the interface magnetization is distinctly different from that of the bulk, resulting in large angular deviations between the two.

Magnetization-induced second-harmonic generation (MSHG) [24–27] is a nonlinear optical version of the magneto-optical Kerr effect (MOKE) technique that provides intrinsic interface sensitivity [28] and a very large magneto-optical response [29]. While MOKE averages the magnetic behavior over the entire film thickness, MSHG very selectively detects *only the interface contribution* due to basic symmetry constraints [28]. We present here detailed MSHG and MOKE studies of the in-plane magnetic anisotropy and switching behavior in Fe/AlGaAs(001) heterostructures. Our results show clearly: (i) the magnetization reversal is strikingly different for the interface and the bulk; (ii) the angle between the interface and bulk magnetization can be as large as 40–85 degrees; and (iii) the ratio of the uniaxial to cubic anisotropy terms $r = K_u/K_1$ is larger at the interface ($|r| \cong 1$) than in the bulk ($|r| = 0.4$). Furthermore, we show for the first time that MSHG is a powerful technique to study interface magnetic

properties in noncentrosymmetric hybrid structures as well as in centrosymmetric systems.

Epitaxial Fe films with 10 nm and 50 nm thicknesses were grown at a substrate temperature of 10–15 °C by MBE on $Al_{0.08}Ga_{0.92}As$ epilayers grown on GaAs(001) substrates [5]. Previous work has shown that the Fe/GaAs(001) interface is essentially abrupt and characterized by Fe-As bonding for samples grown in a similar manner in the same laboratory [11,12]. The 50 nm Fe film was capped with a 5 nm thick Cr layer to prevent surface oxidation. The 10 nm Fe film was exposed to air which results in a considerable reduction of the MSHG signal from the Fe film surface [25]. This is required to measure the magnetization at the Fe/AlGaAs interface with MSHG. The macroscopic magnetic properties were determined by standard MOKE measurements and vibrating sample magnetometry (VSM).

MSHG experiments were performed with a Ti:sapphire amplifier system generating 150 femtosecond pulses with 1 mJ energy at 1 kHz repetition rate and 800 nm wavelength. The attenuated laser beam (15 mW) is focused to a $\sim$500 μm diameter spot on the sample at an angle of incidence of 45°. A small MSHG signal is generated in the direction of the reflected laser beam, and is detected with a high signal-to-noise ratio using a photomultiplier tube and a chopper in combination with a lock-in amplifier. Proper filtering is required to separate the MSHG light from the laser beam. For magnetic anisotropy measurements, the sample is mounted on a computer-controlled rotation stage between the poles of an electromagnet with the magnetic field applied in the plane of the Fe film. The experimental setup allows longitudinal MSHG and MOKE measurements for the same sample orientation.

To the best of our knowledge, this is the first application of MSHG to a noncentrosymmetric system in which a bulk response may be generated. However, this bulk derived signal can be avoided by judicious selection of input- and MSHG-light polarization combination as employed here. Following the approach of Pan, Wei, and Shen [24], it is convenient to separate the MSHG susceptibility into an even (χ^+) and an odd (χ^-) part in the magnetization M: $\chi^{\pm}_{ijk}(-M) = \pm\chi^{\pm}_{ijk}(M)$. Thus the induced MSHG polarization $\vec{P}(2\omega)$ is given by

$$P_i(2\omega) = \chi^{+}_{ijk}(M)E_j(\omega)E_k(\omega) + \chi^{-}_{ijk}(M)E_j(\omega)E_k(\omega), \tag{1}$$

where $\vec{E}(\omega)$ is the local excitation field at frequency ω, and we implicitly assume a summation over the repeated indices. The magnetization sensitive components $\chi^{-}_{ijk}(M)$ arise only from the surface or interface of the Fe film. For s-input polarization and s-polarized MSHG signal, a large bulk response from the AlGaAs (001) substrate can be avoided. For this polarization combination the MSHG signal contains only a response from the longitudinal M_x component. By mixing a small p-polarized contribution to the MSHG signal, M-H loops from the interface can be obtained with very high sensitivity and high signal-to-noise ratio. This polarization combination is utilized for the measurements shown in Figs. 1(a)–1(d).

Figure 1 shows a set of typical MSHG (top panel) and MOKE (bottom panel) magnetization curves for the 10 nm Fe (001) film with the magnetic field applied along the different crystallographic directions indicated. The experimental configuration is shown in Fig. 2(a). In the longitudinal geometry we detect the in-plane component of the magnetization M_x in both the MSHG and MOKE measurements. The most striking difference between MSHG and MOKE M-H loops is the switching behavior of the magnetization reversal process. In the case of the MSHG curves, one-jump switching of the interface magnetization is observed for all principal crystallographic axes [Figs. 1(a)–1(d)]. In contrast, the MOKE M-H loops exhibit distinct plateaus and two switching fields, revealing a two-jump reversal process for the bulk Fe film [Figs. 1(e)–1(h)]. The difference in the magnetization reversal process is particularly apparent for the hard axis [1-10] as shown in Figs. 1(b) and 1(f). These data reveal that the magnetiza-

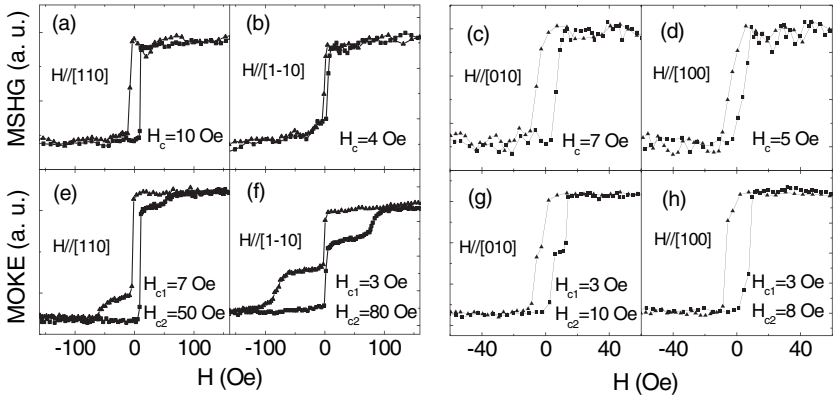

FIG. 1. Longitudinal MSHG, (a)–(d), and MOKE, (e)–(h), M-H loops from 10 nm Fe film with the field applied along the principal crystallographic axes [110], [1-10], [010], and [100], respectively. The squares (triangles) indicate increasing (decreasing) magnetic field. The coercive fields are listed in each panel.

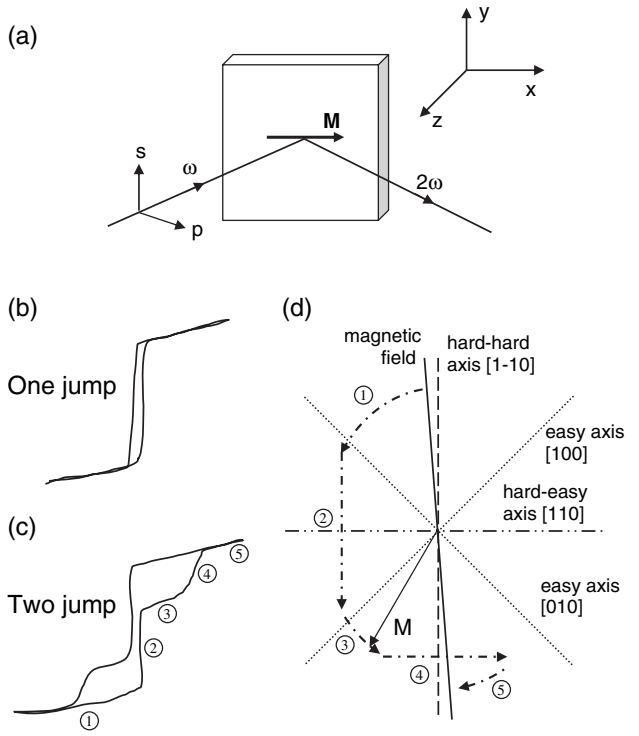

FIG. 2. (a) Experimental configuration, (b) *M-H* loop for one-jump switching; (c) *M-H* loop for two-jump switching, and (d) sequence of the two-jump process. The numbers relate the steps in the sequence to the corresponding feature in the *M-H* loop.

tion of the interface layer switches in a manner which is distinctly different from that of the bulk, and is not rigidly locked to the bulk by the strong exchange coupling typically associated with ferromagnetic metals.

The reversal process of the thickness-averaged (macroscopic) magnetization in the Fe/GaAs system has been studied in detail [19,20], and the mechanisms are summarized in Figs. 2(b)–2(d) using the terminology of Ref. [20]. The ''hard-hard'' axis along [1-10] is the hardest magnetic in-plane axis produced by the UMA, the ''hard-easy'' [110] axis is the intermediate axis, and the [100] and [010] directions are equivalent easy magnetic axes for this film thickness. One-jump switching occurs when M is first pulled over (''jumps'') the hard-hard axis, and thus has sufficient energy to immediately rotate over the hard-easy axis. This produces the conventional looking *M-H* loop of Fig. 2(b). This switching occurs for all crystallographic directions when the uniaxial anisotropy is stronger than the cubic anisotropy, i.e., $|r| \geq 1$ [20]. Hence, our MSHG data show that the anisotropy at the AlGaAs interface is dominated by the UMA contribution.

Two-jump switching [Figs. 2(c) and 2(d)] occurs when M is first pulled over the hard-easy axis, but lacks sufficient energy to cross the hard-hard axis. As the magnitude of the reversed applied field increases further, the second jump occurs when M rotates through this hard-hard axis. These two jumps result in intermediate plateaus or kinks in the *M-H* loop and two distinct switching fields [Fig. 2(c)]. One important point about this mechanism is that it requires two distinct hard axes in the plane of the sample. This condition only arises when the ratio of uniaxial to cubic anisotropy ($r = K_u/K_1$) is less than unity [20]. We determined an anisotropy ratio $|r| = 0.4$ for the bulk magnetization from coherent magnetization precession measurements [30], indicating that the switching processes are in very good agreement with the predictions based on the coherent rotation model [20].

A quantitative analysis of the magnetization curves shown in Figs. 1(b) and 1(f) reveals that the deviation angle between bulk and interface magnetization is in the range $40° - 85°$ between the first and second switching step. This is because the bulk magnetization switches first to the easy [100] axis, as shown in Fig. 2(d), whereas the interface magnetization switches directly to the easy [010] axis. An estimate of the exchange length $l_c = (A/K_1)^{1/2}$ shows that $l_c \approx 20$ nm, where $A = 10^{-11}$ J/m is the exchange stiffness and $K_1 = 4.8 \times 10^4$ J/m^3 is the anisotropy energy density [9]. A micromagnetic calculation which assumes a fixed coupling strength between all the magnetic layer planes, including the interface, shows that the deviation angle between bulk and interface spins would be less than 10 degrees for the 10 nm thick Fe film. Such a model cannot explain the present experimental results.

The assumption of a constant interlayer exchange coupling strength may not be true for layers with different lattice parameters, bonding environment, and magnetic character. An interface layer with different magnetic interaction could dramatically reduce the exchange coupling and cause the abrupt change of switching characteristics. Gordon *et al.* found a body-centered tetragonal distortion in a 9 ML thick Fe film on GaAs(001) [31]. The measured distortion involves an in-plane contraction and an out-of-plane expansion. This structural anisotropy could dramatically affect the magnetic interaction and reduce the exchange coupling normal to the planes. In addition, small variation of the Fe-As bonding may also affect the magnetic character of the Fe interface layer. Fe-As *pd* hybridization has been shown to affect (quench) the Fe magnetic moment and the exchange coupling as illustrated in theoretical calculations [32]. We therefore propose that the specific bonding and structure at the Fe/AlGaAs interface leads to markedly different magnetic anisotropy and exchange coupling of the Fe interface layer with respect to the bulk spins.

For comparison, we utilized the MSHG technique to probe the magnetic anisotropy of the Fe/Cr interface. A 50 nm thick Fe film grown on AlGaAs and capped with a 5 nm Cr layer was used for this experiment. The large Fe thickness eliminates the MSHG signal from the Fe/AlGaAs interface because of the short absorption depth (~ 20 nm) of the MSHG signal at 400 nm wavelength. The *p*-input polarization and *s*-polarized MSHG signal [Figs. 3(a) and 3(b)] provides the best signal-to-noise ratio.

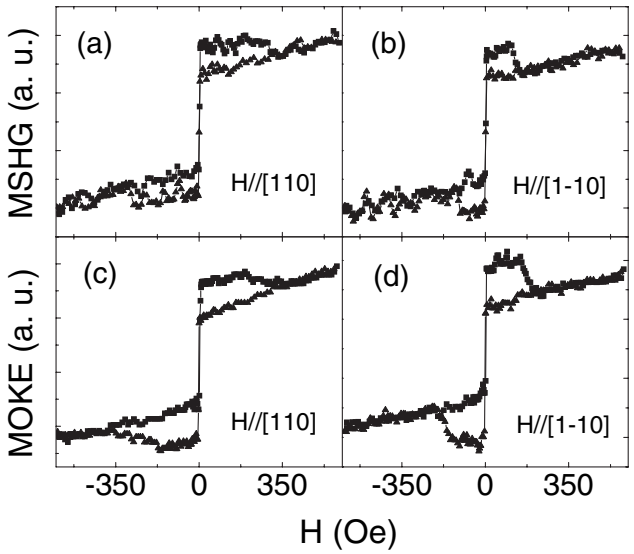

FIG. 3. Longitudinal MSHG, (a) and (b), and MOKE, (c) and (d), *M-H* loops from the 50 nm Fe film capped with a 5 nm Cr layer with the field applied along the hard-easy [110] and hard-hard [1-10] axes, respectively. The squares (triangles) indicate increasing (decreasing) magnetic field.

Figure 3 shows a set of typical longitudinal MSHG (top panel) and MOKE (bottom panel) magnetization curves with the magnetic field applied along the hard $\langle 110 \rangle$ crystallographic directions. Both the MSHG and MOKE *M-H* curves clearly show a two-jump reversal process, in contrast to the switching behavior of the 10 nm Fe film. We determined an anisotropy ratio $|r| < 0.1$ from coherent magnetization precession measurements, indicating a very small UMA contribution at this interface. A two-jump reversal process for this value of $|r|$ is again in very good agreement with the predictions of the coherent rotation model [20]. These results clearly indicate that the Fe/Cr interface contributes a negligible UMA component. The MSHG and MOKE *M-H* loops of Fig. 3 both exhibit an ''overshoot.'' This effect can be attributed to an optical effect caused by ''mixing in'' a small contribution from the transverse magnetization [19].

In conclusion, we have studied the interface magnetic anisotropy and switching behavior in Fe/AlGaAs(001) heterostructures utilizing the longitudinal MSHG technique. We find a pronounced difference in the reversal process of the bulk and interface magnetization—while single step switching occurs at the Fe/AlGaAs interface layer, two-jump switching occurs in the bulk Fe for the magnetic field orientations employed. The angle between the interface and bulk magnetization can be as large as 40°–85°, which is attributed to a decoupling of bulk and interface spins. This occurs as a consequence of the large difference in the magnetic anisotropy ratios arising from interface-induced contributions which we attribute to the specific bonding and structure at the Fe/AlGaAs interface. Our results further show that MSHG is a powerful technique to probe interface magnetic properties in noncentrosymmetric hybrid structures as well as in centrosymmetric systems.

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