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Magnetic Properties of Epitaxial Fe/(Ga,Mn)As Hybrids

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Thin-film structures composed of two kinds of ferromagnetic material — metallic Fe and semiconducting (Ga,Mn)As — were investigated by means of SQUID magnetometry and ferromagnetic resonance spectroscopy. Dependence of remnant magnetic moment on temperature showed unexpected anisotropic features when recorded along two orthogonal in-plane directions. For one of these orientations, the change in sign of the slope of $m(T)$ curve at the Curie point of (Ga,Mn)As was observed, while for the other, an analogous $m(T)$ curve retained monotonic character. Based on the comparison with ferromagnetic resonance data, the apparent non-monotonicity was attributed to the temperature-induced change of balance between the external magnetic field and uniaxial magnetic anisotropy in the plane of Fe layer.

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1. Introduction

The remarkable success of the information technologies as observed in the recent decades was mainly founded on the unprecedented progress in two areas of applied physics: semiconductors and magnetism. While silicon and other semiconductors are omnipresent in computational units, the application of ferromagnetic-metal-based spin valves and tunnel junctions made it possible to widespread the large-capacity memory and storage devices [1, 2]. Thus combination of dilute magnetic semiconductor (DMS) [3] and metallic ferromagnet within a hybrid structure may be seen as a viable alternative in overcoming the limitations of technological applicability of both aforementioned fields, especially if a ferromagnetic metal of high Curie temperature is used. Such hybrid concept may combine the advantages of both types of materials, and — for example — extend the relevant qualities of DMS above its original Curie temperature thanks to the influence of metal's stray fields [4, 5].

2. Experimental details

The ferromagnetic heterostructures studied in this paper were grown in a Riber 32 R&D molecular beam epitaxy (MBE) system on a semi-insulating GaAs wafers of [001] orientation. Immediately on the substrate a 100 nm thick buffer of high-temperature GaAs was deposited at 600 °C, followed by a 2 nm thick buffer of low temperature GaAs and a 100 nm thick film of ferromagnetic (Ga,Mn)As, both grown at 250 °C. The condition of low substrate temperature was exercised in order to introduce about 5% of manganese ions into the lattice. The wafer was then transferred to another chamber via an ultrahigh vacuum channel and without exposure to the atmosphere. There a Fe layer of different thickness in the range of 3–11 nm was deposited directly from an e-beam evaporator on the (Ga,Mn)As surface at room temperature. Finally, such hybrid samples were capped with a 2 nm thin Al film to prevent oxidation of iron. For the whole series, a deposition process was monitored *in situ* with use of the reflection high-energy electron diffraction technique that confirmed good crystalline quality of both (Ga,Mn)As and Fe constituents.

Magnetometric characterization was performed with Quantum Design XL7 high sensitivity superconducting quantum interferometer (SQUID) operating for up to 7 T in a range of temperatures from 2 to 400 K. Both the hysteresis loops and temperature-dependent curves of remnant magnetic moment were recorded along different crystallographic directions. Ferromagnetic resonance (FMR) experiments were carried out with use of a Bruker electron spin resonance spectrometer working at X-band microwave frequency (≈ 9.4 GHz) and equipped with a continuous-flow helium cryostat that enabled temperature-dependent measurement in the range of 5–300 K. When needed, a complete anisotropy of FMR was recorded, with the external magnetic field $\boldsymbol{H}$ varied in three orthogonal planes of (Ga,Mn)As: (110), $(1\bar{1}0)$, and the (001) growth plane.

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3. Results and discussion

Figure 1 shows dependence of remnant magnetic moment on temperature for two representative

Fe/(Ga,Mn)As heterostructures with different thickness of the metallic part. In both cases a well known two-component curve is featured, with distinct Curie transition of (Ga,Mn)As at $T_{\mathrm{C}} \approx 50$ K. It is worth mentioning that among the whole set of four studied Fe/(Ga,Mn)As samples, which (Ga,Mn)As layers were grown under nominally the same conditions, in each and every case the revealed Curie temperature of (Ga,Mn)As was about 50 K, thus confirming good control of the fabrication process. Under assumption that magnetization of Fe layers at 300 K is equal the bulk value of 1700 emu/cm^3 [6], the thicknesses of Fe film in different samples were estimated to be in the range of 2.9–10.8 nm.

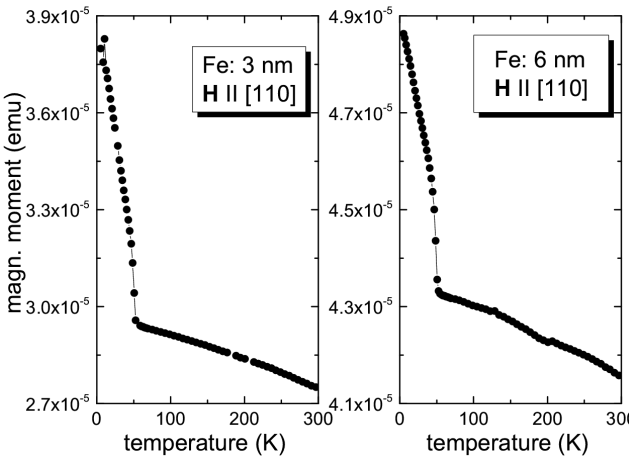

Fig. 1. Magnetic moment versus temperature for Fe/(Ga,Mn)As samples with different thickness of Fe layer (3 and 6 nm), as measured along [110] axis of (Ga,Mn)As.

Dependence of remnant magnetization on temperature changes substantially when the measurement axis (i.e. the orientation of small magnetic field of 50 Oe) is rotated by 90 degrees from [110] to [$1\bar{1}0$]; see Fig. 2. Now the m vs. T curve is no longer monotonic and the sign of its slope changes at T_{C} of (Ga,Mn)As. The change in sign of the slope is to some extent resemblant to the features expected in double layers with antiferromagnetic interfacial coupling (excluding high-temperature drop of the net magnetic polarization) [7]. Such interaction [NewRef1] and a cusp of m vs. T curve [NewRef2] have been indeed reported for Fe/(Ga,Mn)As system. But since the interfacial coupling does not vanish only when (Ga,Mn)As remains ferromagnetic and since the magnetization of Fe changes insignificantly at low temperatures, at TC of (Ga,Mn)As one may only expect the maximum of net magnetic moment of Fe/(Ga,Mn)As hybrid. Moreover, the cited observation by Olejnik et al. was made under zero field cooling conditions. The results presented here reveal qualitatively different shape of m vs. T curve, with no effect of (zero) field cooling. The another signature of antiferromagnetic interaction, i.e. the exchange bias shift of the hysteresis loop, was not found either, thus one may conclude on the absence of interfacial coupling in the samples of this study. Even more complex features were observed for this orientation of $\boldsymbol{H}$ in hysteresis loop measurements, as exemplified in Fig. 3. Although comprehensive explanation for these multi-loop shapes require further detailed study that reaches beyond the scope of this short communication, it is already possible to identify contributions from Fe and (Ga,Mn)As by a comparison of data collected at 300 K and 5 K.

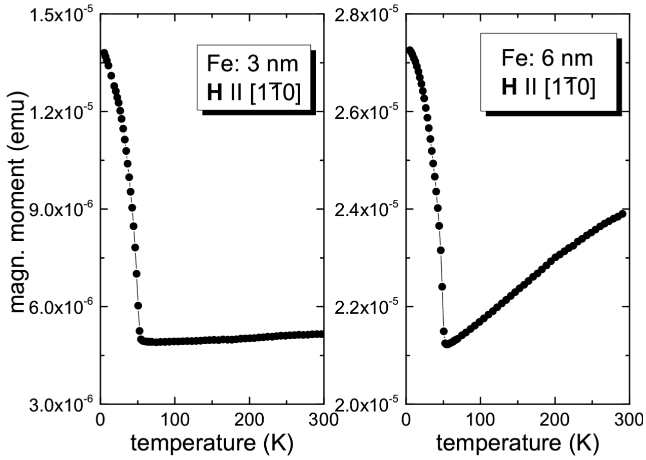

Fig. 2. As in Fig. 1, but along [$1\bar{1}0$] axis of (Ga,Mn)As.

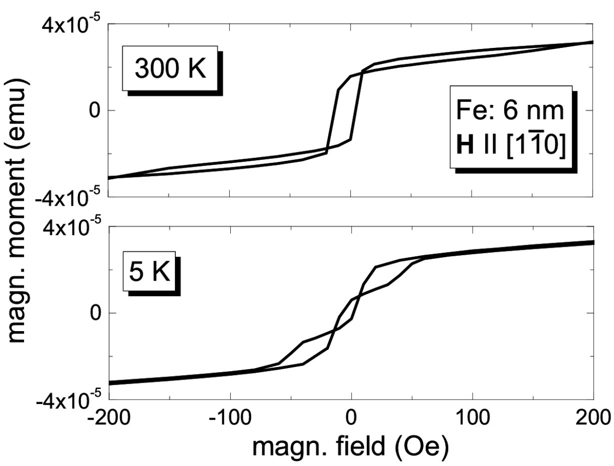

Fig. 3. Hysteresis loops of Fe/(Ga,Mn)As heterostructure with 6 nm of Fe, measured at different temperatures along [$1\bar{1}0$] axis of (Ga,Mn)As.

In order to elucidate the above mentioned findings of magnetometric studies, the angle-dependent ferromagnetic resonance measurements were performed. In the polar configuration, i.e. with the external magnetic field varied between normal and in-plane orientations, the familiar uniaxial behavior was recorded, as depicted in Fig. 4. The minima of resonance fields — and so the orientations of the magnetization easy axes — were for both materials found in the sample plane, which is a typical demagnetization anisotropy of thin-film-shaped ferromagnets [10]. In the case of epitaxial (Ga,Mn)As, such uniaxial pattern is also attributed to the tensile tetragonal distortion along the growth axis [11]. For some of the studied heterostructures, well resolved multi-line FMR features were observed, a manifestation of non-uniform (spin wave) collective excitations of the spin system [12].

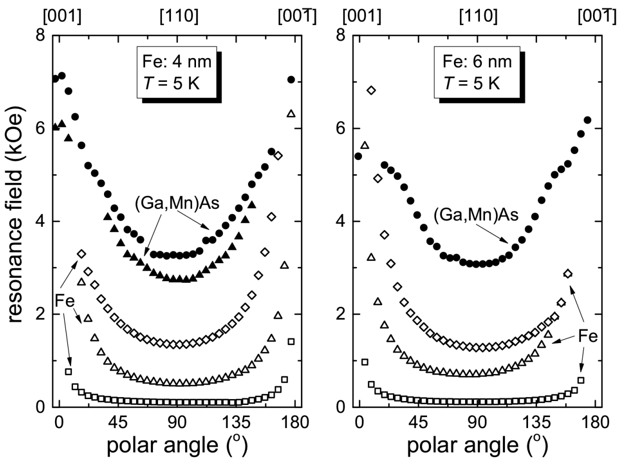

Fig. 4. FMR resonance fields collected in the out-of--plane (polar) configuration for Fe/(Ga,Mn)As samples with 4 nm (left) and 6 nm (right) of Fe. In top of the graph, the corresponding crystallographic directions of (Ga,Mn)As are depicted. Multi-line features are the manifestation of non-uniform (spin wave) excitations. For (Ga,Mn)As the top, stronger line was identified as the uniform FMR mode, whereas for Fe the dominating, uniform-mode-related line was observed at the lowest fields.

Finally, the results of in-plane FMR measurements are presented in Fig. 5, with clear distinction between resonance fields corresponding to (Ga,Mn)As and Fe components. In the former case, the superposition of cubic and uniaxial anisotropies was revealed, a typical feature of ferromagnetic (Ga,Mn)As layers [13]. For Fe, on the other hand, pure uniaxial in-plane anisotropy was identified, which is somewhat surprising, since in prior in-plane FMR studies of Fe layers grown on nonmagnetic GaAs the strong biaxial symmetry had been observed with only limited contribution of the uniaxial anisotropy (see inset of Fig. 5). Nevertheless, the manifestation of uniaxial--only type of in-plane magnetic anisotropy in Fe being part of Fe/(Ga,Mn)As structure, brings an explanation for the above described non-monotonicity of low-field m vs. T curves.

When the external magnetic field is applied along the magnetization easy axis of Fe, that is [110] direction of (Ga,Mn)As, then for any temperature exercised in this study the Fe magnetic moment remains parallel to $\boldsymbol{H}$. In such a case, the signal measured by SQUID is a simple sum of Fe magnetic moment and a projection

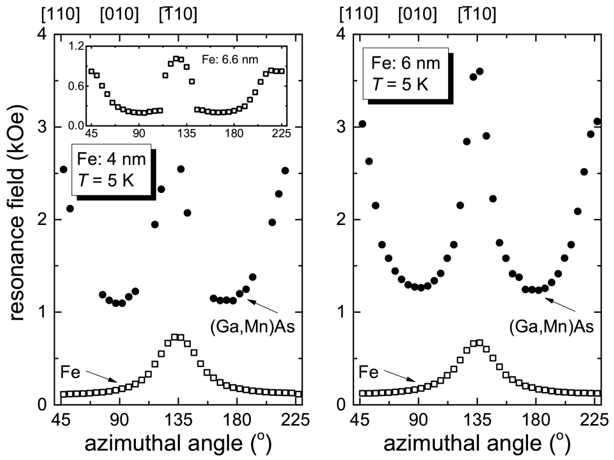

Fig. 5. FMR resonance fields collected in the in-plane (azimuthal) configuration for Fe/(Ga,Mn)As samples with 4 nm (left) and 6 nm (right) of Fe. In top of the graph, the corresponding crystallographic directions of (Ga,Mn)As are depicted. Inset: azimuthal FMR anisotropy observed in a reference 6.6 nm thin film of Fe grown on non-magnetic GaAs. Units are the same as in the main graph parts.

of (Ga,Mn)As magnetic moment onto [110] direction†. On the other hand, when the magnetic field is rotated by 90° to [1$\bar{1}$0] direction of (Ga,Mn)As, which is the in--plane hard axis for Fe, one can expect a competition between the applied low magnetic field and the uniaxial anisotropy. The former attempts to bend the Fe magnetic moment towards [1$\bar{1}$0], but the latter tends to align it with [110]. With decreasing temperature, the absolute values of magnetocrystalline anisotropy constants of Fe increase [14], and the effect of magnetic anisotropy gets stronger as compared with the influence of magnetic field applied. Thus for temperature swept from 300 K down to T_C of (Ga,Mn)As, diminishing projection of Fe magnetic moment on the direction of $\boldsymbol{H}$ is observed. When temperature is lowered further down to 5 K, the dominant effect of increasing magnetic polarization in thick (Ga,Mn)As layer overshadows the contribution of thin Fe.

4. Conclusions

The novel ferromagnetic hybrids consisting of (Ga,Mn)As and Fe had been successfully grown by means of the molecular beam epitaxy and subsequently investigated with use of SQUID and FMR techniques. Both magnetometric and microwave spectroscopy data provided evidence for two distinct ferromagnets. The analysis of differing symmetries of in-plane magnetic anisotropies in both components facilitated explanation for the unexpected observation of non-monotonic

†It should be reminded here that [110] is not the easy axis of (Ga,Mn)As

dependence of hybrids' remanence magnetic moment on temperature. Still some unresolved issues await explanation, including dissimilar azimuthal anisotropy of FMR in Fe when grown on (Ga,Mn)As and GaAs. One should bear in mind that in present study the heterostructures of different Fe thicknesses were grown in separate MBE processes. Thus for even better quality control, the initial successful attempts were made in MBE growth of Fe/(Ga,Mn)As with varying Fe thickness, and the preliminary X-ray diffraction data revealed about 5–6 nm change in Fe layer thickness across the sample. Further experimental studies of Fe/(Ga,Mn)As hybrids with Fe gradient will follow.

Acknowledgments

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