

Adjustable anisotropy in ferromagnetic (Ga,Mn) (As,P) layered alloys

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We show that the extension of the widely studied GaMnAs system to quaternary $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ alloys allows to change and inverse the Mn doping induced uniaxial strain and thus the uniaxial magnetocrystalline anisotropy fields. In particular for typical Mn concentrations of $x=0.07$ the Mn dopant induced strain can be completely compensated by phosphorous alloying in the $y=0.06$ range and layers with both in-plane or out-of-plane easy axes of magnetizations with extremely low barriers for magnetization switching can be obtained.

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Diluted ferromagnetic (III,Mn)V semiconductors such as GaMnAs have the unique property that contrary to conventional metallic ferromagnets the shape anisotropy is weak as compared uniaxial strain related anisotropies. Further, the concentrations of the free carriers, mediating the ferromagnetic interactions, are much lower in the 10^{20} cm^{-3} range—which allows to manipulate them by external perturbations. Whereas this offers *a priori* the possibility to Taylor ferromagnetic thin layers with different anisotropies the close relation with the Mn doping concentration limits such attempts strongly. Generally, GaMnAs films are grown by low-temperature molecular beam epitaxy (LTMBE) on GaAs substrates. Due to strain induced by the lattice mismatch between the GaMnAs epitaxial film and the underlying substrate anisotropy fields of several kOe are encountered.^{1–4} Recently a different growth technique has also been proposed: it is based on ion implantation followed by pulsed laser melting (IPLPM).^{5,6} However the structural and magnetic properties of the LTMBE and IPLM grown samples are not the same; IPLM samples are characterized by residual electrical compensation, inhomogeneous dopant distribution and potentially by second phase inclusions. For device application it is interesting to control the magnetic anisotropy and create conditions for easy switching of the magnetization by applied electrical or magnetic fields or by spin currents. Various techniques have been proposed to manipulate the anisotropy independently from the Mn doping concentration. Examples are carrier depletion in diode structures by the application of an external electric field⁷ or the use of multilayers structures composed of GaMnAs and piezoelectric⁸ or ferroelectric top layers.⁹ Such manipulations would be more effective if smaller anisotropies and smaller carrier concentrations could be achieved. We show here that it is possible to overcome this bottleneck by alloying with phosphorous. First results have already been reported recently.^{4,5,10,11} In this letter, we report the results of an extensive study of the strain, electrical properties and magnetic anisotropies of LTMBE grown GaMnAsP quaternary thin films which show that it is possible to combine high Mn doping levels required for robust ferromagnetism with small barriers for magnetization switching.

50-nm-thick $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ films were grown by low-temperature molecular beam epitaxy on GaAs (100) sub-

strates. The concentration of phosphorous was varied from $y=0$ to $y=0.08$ and the Mn concentration was set to $x \approx 0.07$. The P concentrations were determined from the lattice constants of a series of GaAsP reference samples grown at the same conditions.¹⁰ After the growth the samples were thermally annealed under N_2 atmosphere at 250° for 1 h. The magnetic properties of the films were characterized by magnetometry using a superconducting quantum interference device and ferromagnetic resonance (FMR) spectroscopy. The uniaxial strain ε_{zz} defined as $\varepsilon_{zz}=(a_{\perp}-a_{\text{rel}})/a_{\text{rel}}$ (Ref. 3) was determined by high resolution x-ray diffraction. The magnetocrystalline anisotropy constants and associated anisotropy fields were determined by X band (9 GHz) FMR spectrometry. The angular dependence of the $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ FMR spectra were measured in two planes, (001) and (110). These two sets of measurements enable us to determine the resonance position for the high symmetry orientation of $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ film: $\text{H} \parallel [001], [110], [1-10]$ and $[100]$ from which the anisotropy constants can be determined.^{2,12}

The sheet resistivity of the series of $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ samples containing different concentrations of P is shown in Fig. 1. The samples were taken from different growth runs with very similar Mn concentrations. They all present very similar critical temperatures between 110 and 135 K. We see that the conductivity of the samples is metallic in all cases as expected for a Mn doping of $x \approx 0.07$ but varies within a

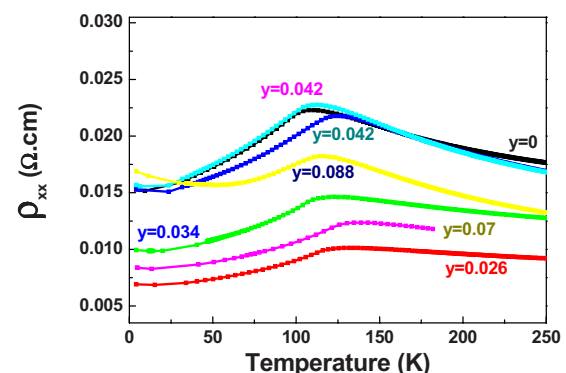


FIG. 1. (Color online) Electrical resistivity of the different $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ samples as a function of temperature.

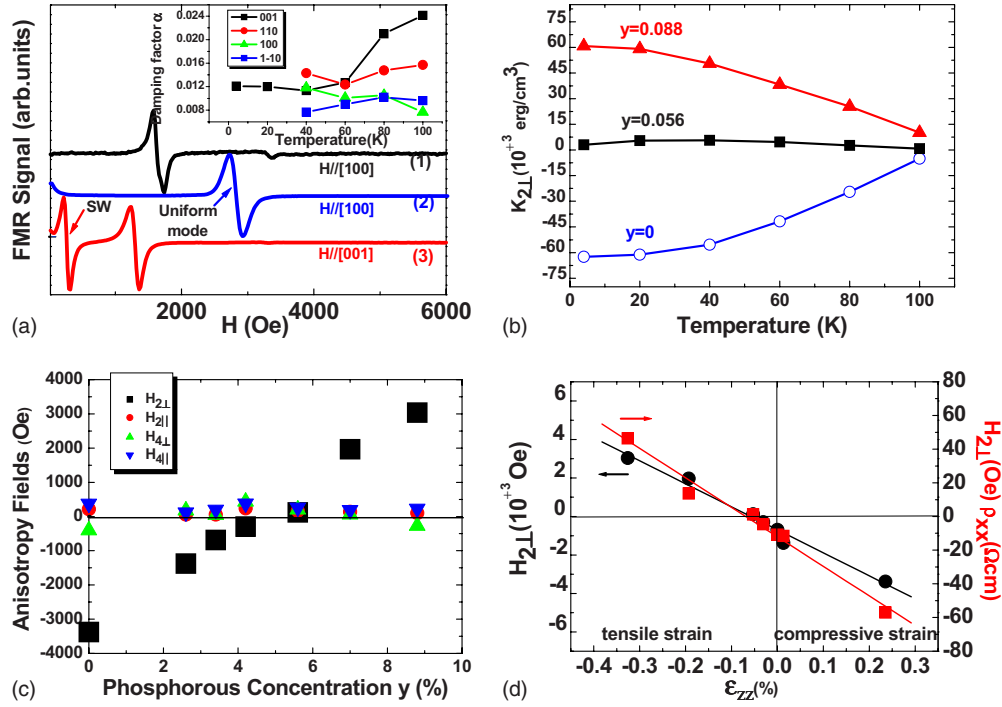


FIG. 2. (Color online) (a) FMR spectra of $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ 50-nm-thick annealed samples for applied fields along the easy axis; the easy axis of magnetization is parallel to $[100]$ for the samples $y=0$ (1, black), $y=0.056$ (2, blue) and has switched to out-of-plane, i.e., $\parallel[001]$, for the sample $y=0.088$ (3, red). Inset: sample $y=0.088$; damping factor as a function of temperature and magnetic field orientation; $H\parallel[001]$ squares (black), $H\parallel[110]$ up triangles (green), $H\parallel[100]$ circles (red), $H\parallel[1-10]$, and down triangles (blue). (b) The magnetocrystalline anisotropy constants as a function of temperature for the samples $y=0$ (blue), 0.056 (black), and 0.088 (red). The lines are guides for the eyes. (c) The magnetic anisotropy fields at 4 K as a function the phosphorous concentration. (d) The anisotropy fields $H_{2\perp}$ at 4 K before (black) and after normalization (red) for a constant hole concentration as a function of strain ϵ_{zz} . Error bars for the anisotropy constants and fields are estimated to $<5\%$ which is below the size of the symbols

factor of three. The weak dependence of the T_c ($\sim p^{1/3}$) which is predicted by the mean-field model^{13,14} explains that the Curie temperature in this series varies nevertheless only slightly. In principle we should expect a systematic variation in the hole concentration with the phosphorous content as the acceptor level becomes increasingly deeper. However, the Mn incorporation is also influenced by the P alloying and more detailed measurements are required in order to investigate this interdependence.

Figure 2(a) shows typical FMR spectra of the $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ samples with concentrations of P $y=0$, 0.056 , and 0.088 . The applied magnetic field is in each case aligned parallel to the respective easy axes. For phosphorous concentrations $y=0$ to $y=0.056$ the films have an easy in-plane axis along $[100]$, with resonance fields near (1) $H=1632$ Oe, (2) $H=2831$ Oe, and hard axes along the $[001]$ direction $H=7816$ Oe, $H=3761$ Oe (not shown here). This corresponds to the case of compressively strained films on GaAs substrates. For higher concentrations of phosphorus we observe a change in the easy axis of magnetization to $[001]$ i.e., perpendicular to the film plane. The dominant uniaxial anisotropy constant $K_{2\perp}$ can be continuously varied from highly negative to highly positive values by adjusting the P alloying level. The FMR spectra of the highly P doped samples show in addition to the uniform mode (Fig. 2(a)) two spin wave resonances with resonance fields near $H=1289$ and 266 Oe, respectively. The hard axis of the sample

with $y=0.088$ was observed to be along the $[110]$ direction, $H=4977$ Oe (not shown here).

Analyzing the FMR spectra via the Smit-Beljers formalism¹⁵ and taking into account the minimization of the free energy for the equilibrium states of the magnetization we have determined the four magnetic anisotropy constants $K_{2\perp}$, $K_{2\parallel}$, $K_{4\perp}$, and $K_{4\parallel}$ which describe the biaxial and cubic anisotropies. As it has been shown previously,² the dominant constant for GaMnAs layers on GaAs or GaInAs substrates with typical Mn concentrations of $x=0.07$ is $K_{2\perp}$ which is proportional to the uniaxial strain induced by the lattice mismatch. Its value at low temperature and its variation with temperature are shown in Fig. 2(b) for three samples representing the cases of high compressive, high tensile, and close to zero strain. These cases correspond to P concentrations of $y=0$, 0.088 , and 0.056 . For the $y=0$ sample the $K_{2\perp}$ constant at $T=4$ K has a negative value of -6.10^4 erg/cm³; with increasing P doping level its value increases monotonously and passes through zero (for $y=0.056$). For this range of phosphorous concentration the four anisotropy constants are small and of comparable numerical value. With further increase in the (P) $K_{2\perp}$ becomes positive and the easy axis of magnetization switches to out-of-plane. In Fig. 2(c) we show the anisotropy fields 2 K/M as a function of phosphorous concentration. The fact of dividing the anisotropy constant by the magnetization allows us to correct the variation in $K_{2\perp}(y)$ for potentially slightly different Mn concentra-

tions. In a previous publication we have experimentally confirmed that the $K_{2\perp}$ constant depends linearly on the carrier concentration and the uniaxial strain.¹² Assuming that the mobility is similar for all samples of this series we can take into account slightly different hole concentrations from the conductivity values and we then expect a linear variation in the anisotropy fields normalized to a constant hole concentration with the uniaxial strain. Indeed, this is observed in the Fig. 2(d). In this figure the anisotropy field $H_{2\perp}$ at 4 K and the its normalized value ($H_{2\perp} \cdot \rho_{xx}$) are plotted as a function of the uniaxial deformation ε_{zz} . The linear variation is in good agreement with the mean field model, as it has been shown also for the case of GaMnAs.¹² Our results demonstrate that it is possible to manipulate the uniaxial anisotropy field to any desired value just by varying the P doping level without any loss of sample quality.

It can be seen in Fig. 2(d) that the linear fit to these results does not pass through the origin. Whereas such behavior would have been expected for the total perpendicular anisotropy field due to the contribution of the demagnetization field this is not the case for the $H_{2\perp}$ defined by $2K_{2\perp}/M$. We describe this small deviation to the fact that the samples have also slightly different values of the exchange splitting parameters due to the alloying which will scatter the corresponding $K_{2\perp}$ values.³

The observation of spin-wave resonances in addition to the uniform mode spectrum in highly P doped samples gives us important information on the physics in these quaternary alloys. It allows us to determine the effective exchange integral between the Mn ions; according to theoretical predictions its value is expected to be increased relative to the case of GaMnAs.¹⁶ In order to deduce this parameter we have applied the phenomenological model proposed by Liu *et al.* for [001] and [100] directions of the applied field.¹⁷ The exchange constant, D , deduced at $T=40$ K is ~ 2.733 T nm² which by considering the magnetization value at this temperature leads to the spin stiffness, $A = 0.06$ pJ/m. This value is slightly larger than what has been observed in GaMnAs/GaInAs ($x=0.07$) samples via magneto-optical Kerr effect microscopy,¹⁸ from this value we deduce an effective exchange integral between the Mn ions of $J_{\text{Mn-Mn}} = 0.22$ meV. Note that in contradiction to theoretical predictions the effective exchange integral has not increased considerably by the P alloying.

FMR measurements have been equally performed at a second microwave frequency Q band (35 GHz) in order to determine the Gilbert damping factor from the variation in the uniform mode line width with the microwave frequency. As shown in the inset of Fig. 2 the damping factor of the sample with highest P concentration is $\alpha = 0.012$ at low temperature; this value is about three times higher than the one reported previously for phosphorous free GaMnAs layers.¹⁹ The damping factor varies only weakly with temperature between 4 K and $0.8T_C$ and increases as is usually observed when the temperature approaches T_C .

A further interesting aspect is the small value of the inhomogeneous part of the FMR linewidth. Its value is only 40 Oe which is comparable to the best state of the art GaMnAs samples grown on GaAs(001) substrates. It is however an order of magnitude smaller than that reported in the case of

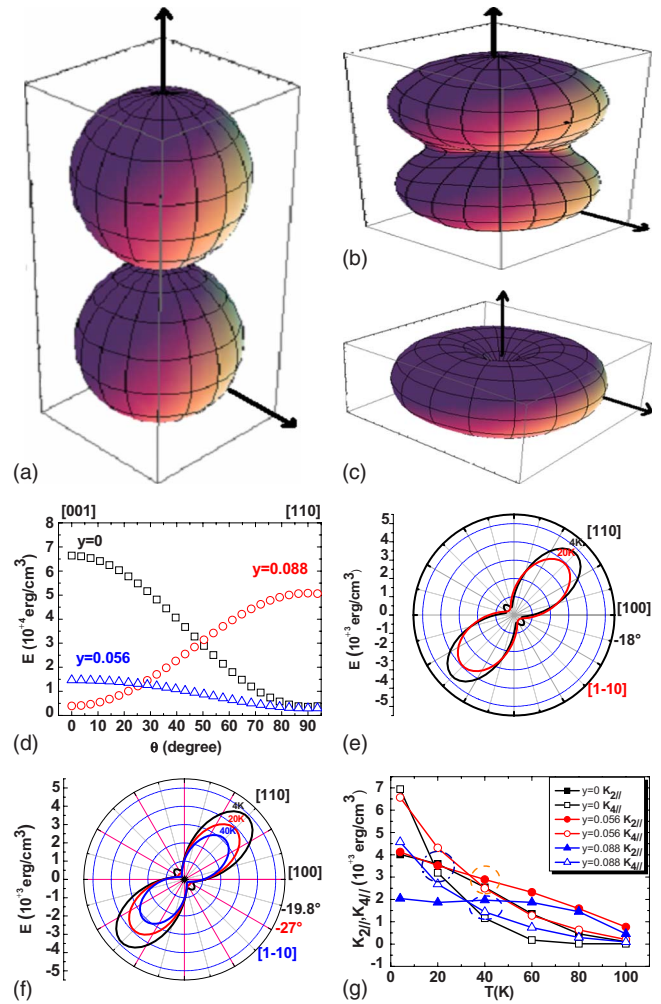


FIG. 3. (Color online) Three-dimensional plot of free-energy densities for $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ at 4 K with (a) $y=0$, (b) 0.056, and (c) 0.088. (d) The energy density for samples $y=0$ (black), 0.056 (blue), and 0.088 (red) as a function of the out of plane orientation θ for $\varphi=45^\circ$. (e) Free-energy densities as a function of the in-plane magnetization orientation $|M||\cos \varphi, \sin \varphi, 0|$ for sample $y=0$ at $T=4$ K (black) and $T=20$ K (red) and (f) for sample $y=0.056$ at $T=4$ K (black), 20 K (red), 40 K (blue). The directions of the energy minima shift toward (Refs. [1–10]) with increasing temperature. (g) The magnetocrystalline anisotropy constants $K_{2\parallel}$ and $K_{4\parallel}$ as a function of temperature for the samples $y=0$, 0.056, and 0.088. The lines are guides for the eyes. The change in easy axes is indicated by the circles.

GaMnAs layers with perpendicular anisotropy, which had been obtained by epitaxial growth on GaInAs substrates (the tensile strained samples). The inhomogeneous linewidth is a measure of the magnetic homogeneity of such films and its low value in phosphorous alloyed films manifests the absence of dislocation related inhomogeneous dopant and charge carrier distributions which are a major drawback for the use of GaInAs buffer layers.

From the numerical values of the anisotropy constants we can calculate the free-energy density E as a function of the crystalline orientations (θ, ϕ) . In Figs. 3(a)–3(c) we show such surfaces for zero applied magnetic field for the three prototype samples with $y=0$, 0.056, and 0.088, respectively.

These surfaces show the energy barriers for the magnetization reorientation and define the preferential axes of the magnetization. A more detailed view can be obtained from the cross sections of the surface in particular planes. For example in Fig. 3(d) we have plotted the out-of-plane angular variation in the free-energy density as a function of θ angle. These figures illustrate that while for the two extreme cases the difference between the energy along the easy and hard axes are considerably large, for the intermediate case of the sample with $y=0.056$ the variation in the energy becomes very small.

This is a very important result as it indicates that one can reduce the energy barriers of the magnetization switching for both out-of-plane and in-plane easy axis configurations. This makes such layers interesting objects in FM/NM/FM trilayer structures currently studied for their tunneling magnetoresistance effects.

The energy surfaces for the in-plane orientations ($\theta=90$) of the magnetization for samples with $y=0$ and 0.056 are shown in Figs. 3(e) and 3(f), respectively. At low temperatures the easy axis of magnetization is not aligned in the absence of applied magnetic fields along any high-symmetry direction due to competing second order and fourth-order anisotropy fields. At $T=4$ K it is oriented along the intermediate direction $\varphi=-18^\circ$ and -19.8° with respect to the $[100]$ direction for $y=0$ and 0.056 , respectively.

When we raise the temperatures to $T=20$ and 40 K respectively the easy axis monotonously shifts to the (Refs. 1–10) direction. These particular orientations correspond to the predominance of the $K_{2\parallel}$ and $K_{4\parallel}$ constants, as shown in Fig. 3(g). Thus the temperature is another parameter in addition to the strain which can be used to manipulate the orientation of the magnetization of $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ ferromagnetic films with a fixed Mn concentration.^{12,20}

In conclusion we have shown that ferromagnetic $\text{Ga}_{1-x}\text{Mn}_x\text{As}_{1-y}\text{P}_y$ layers with $x\approx 0.07$ and $y=0$ to 0.088 of high structural and magnetic qualities can be grown by low-temperature molecular beam epitaxy. Contrary to previous conclusions⁵ the conductivity and critical temperature are only weakly influenced by the phosphorous alloying. For phosphorous doping levels around $y=0.06$ layers with in plane or out of plane easy axes with small barriers for magnetization switching can be obtained. Strain manipulation in quaternary III-V alloys is a promising technique for tailoring spintronics devices.

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