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Observation of Giant Magnetic Linear Dichroism in (Ga, Mn)As

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Giant magnetic linear dichroism (MLD) is observed in the ferromagnetic semiconductor $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$. The contribution to this effect induced by the spontaneous magnetization can be clearly identified by azimuthal dependencies. The spectral dependence of the effect in the range from 1.4 to 2.4 eV shows that the MLD induced by the spontaneous magnetization is strongly enhanced for excitations from the electronic states that are responsible for the ferromagnetism in this material. This spectral sensitivity and the size of the effect makes MLD a powerful tool for the study of (III, Mn)V alloys and similar novel ferromagnetic semiconductors.

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Incorporating a small amount of Mn ions into a semiconductor host results in a giant Zeeman splitting caused by the exchange $sp-d$ interaction between carriers and localized Mn spins [1]. In (III, Mn)V diluted magnetic semiconductors (DMS), the manganese ions act as acceptors [2,3], providing itinerant holes [4] and modifying the valence-band structure [5–7]. These materials reveal strong ferromagnetism [8], currently reaching Curie temperatures up to $T_C = 159$ K, as reported for annealed (Ga, Mn)As epilayers grown at low-temperature conditions [9]. The realization of ferromagnetism in a semiconductor results in many unique phenomena which have no analogy in the standard physics of either magnetism or semiconductors, such as, e.g., the electric field control of ferromagnetic properties of (III, Mn)V structures [10]. It also leads to multifunctionality of DMS-based devices combining different phenomena in one circuit [10–16].

While these novel ferromagnetic semiconductors have attracted intense research interest, the physics of these materials is still subject to debate. One of the controversial issues is the role of the valence-band states. Various approaches towards describing the ferromagnetism in III–V semiconductors have been proposed [17–23], but because of ambiguities in the experiments it has been difficult to pin down the correct description.

One of the most popular optical methods to study and demonstrate the magnetic properties of zinc-blende semiconductors is based on the measurements of their photoluminescence [24], as recombination of spin-polarized carriers leads to circular polarized radiation. However, this method is impractical for (III, Mn)V alloys that hardly show any detectable photoluminescence. This is due to the large number of defects that appear because of the low-temperature growth conditions and act as nonradiative decay centers. Another frequently used technique to investigate magnetic properties of media is based on measurements of magneto-optical phenomena, such as magnetic

circular dichroism (MCD) and Faraday or Kerr effects, that are of first order in the magnetization. There were several attempts to apply these techniques for the study of the electronic structure of (III, Mn)V semiconductors, but the interpretation of the magneto-optical spectra of (Ga, Mn)As appeared to be a complicated problem [25–28]. Moreover, the $sp-d$ interaction in Mn-doped semiconductors is responsible for a strong magneto-optical signal already in the paramagnetic phase [29], so that the ferromagnetic order introduces only small changes in the spectra. Only recently, thorough and systematic studies of MCD in (Ga, Mn)As over a broad spectral and thermal range allowed one to indicate the ferromagnetism related electronic excitations in this particular material [27]. Thus, it is clear that for progress in our understanding of the electronic and magnetic properties of the (III, Mn)V alloys and similar novel ferromagnetic semiconductors, alternative techniques are required.

In this Letter we report the observation of a giant magnetic linear dichroism (MLD) in (Ga, Mn)As which is drastically enhanced in comparison with previous MLD data on this material [30] and in II–VI diluted magnetic semiconductors [31]. Spectral studies of the giant MLD in (Ga, Mn)As show that, in contrast to MCD, this effect is predominantly sensitive to the specific electronic states in the valence band that are responsible for the ferromagnetism. We use (Ga, Mn)As as a demonstrator material to show that MLD, which is normally ignored, can be a powerful technique to study hole-mediated ferromagnetism.

Magnetic linear dichroism is the dichroism of linearly polarized light induced in a medium by an external magnetic field or magnetization [32]. The effect originates from the difference of the optical absorption indices Δk for light linearly polarized parallel and perpendicular to magnetization \mathbf{M} . For a cubic crystal such as (Ga, Mn)As it can be shown that [33]

$$\Delta k = k_{\parallel} - k_{\perp} \propto \mathbf{M}^2. \quad (1)$$

For materials with ferromagnetic order, one can write $\mathbf{M} = \mathbf{M}_{sp} + \gamma\mathbf{H}$, where \mathbf{M}_{sp} is the spontaneous magnetization, γ is the paramagnetic susceptibility of the medium and \mathbf{H} is the external magnetic field. Because of magnetic anisotropy the spontaneous magnetization of a ferromagnet may have an angle with respect to the external magnetic field, which allows one to distinguish several contributions to the MLD. The contribution proportional to M_{sp}^2 can be observed only below the Curie temperature and characterizes the ferromagnetic state of the medium. The second contribution, proportional to $(\gamma H)^2$, can be observed in both paramagnetic and ferromagnetic phases. In addition, there is a cross term, proportional to $\gamma M_{sp} H$.

For the study of the MLD we used a laser polarimeter ($\lambda = 815$ nm) in reflection geometry at near normal incidence, employing a Wollaston prism and a split-diode detector. The external magnetic field was applied in the plane of the sample, slightly off the $[1\bar{1}0]$ crystallographic direction of (Ga, Mn)As [see Fig. 1(a)]. The MLD in this scheme resulted in a different reflection coefficient for light polarized parallel and perpendicular to the magneti-

zation \mathbf{M} in the magnetic layer. This difference was observed as a rotation of the polarization of the reflected light. In addition, we have also performed spectral measurements in the range from 1.4 to 2.4 eV in a conventional scheme with a photoelastic modulator [32], using a halogen lamp as a light source and a monochromator.

The ferromagnetic semiconductor $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$ alloy of high quality, as confirmed by x-ray diffraction [34], was fabricated by molecular beam epitaxy on a (001)GaAs substrate covered by a $3 \mu\text{m}$ $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ buffer layer. The buffer was grown at a temperature $T = 620^\circ\text{C}$, while the 350 nm thick (Ga, Mn)As layer was grown at $T = 270^\circ\text{C}$. Magnetic characterization of the sample using a superconducting quantum interference device magnetometer revealed a Curie temperature of about 50 K and a magnetic anisotropy with two in-plane easy axes along the $[100]$ and $[010]$ crystallographic directions in (Ga, Mn)As, respectively.

A typical field dependence of the MLD in (Ga, Mn)As measured at a photon energy of 1.52 eV is shown in Fig. 1(b) for different angles between the incident polarization and the $[100]$ crystallographic direction in (Ga, Mn)As. One can see that the field dependence is extremely sensitive to the orientation of the incoming polarization. The polarization rotation has a maximum amplitude for $\theta = 45^\circ$ and $\theta = 135^\circ$. At these orientations of the incoming polarization the field dependence is characterized by an M-shaped hysteresis loop with jumplike changes of the signal at $H_{12} = 9$ mT and $H_{23} = 110$ mT, which is illustrated by the $\theta = 137^\circ$ and $\theta = 33^\circ$ curves in Fig. 1(b). Such hysteresis form is typical for the magneto-resistance in (Ga, Mn)As [12,15], as well as for other effects that are even in magnetization and originate from the fourfold magnetic anisotropy of this material. The process of magnetization reversal in an external magnetic field happens via jumps of the magnetization over 90° between the four easy axes directions [indicated as (1) to (4) in Fig. 1(a)], so that in a relatively weak field, the magnetization is practically parallel to one of the $[100]$ or $[010]$ easy axes [12,15,16,30,35].

The shape of the magneto-optical response changes drastically when the polarization of the incident light is parallel to the $[100]$ or $[010]$ crystallographic directions in (Ga, Mn)As. Thus, if θ is close to 0° or 90° , the M-shaped hysteresis is not observed anymore, as can be already seen from the $\theta = 107^\circ$ curve in Fig. 1(b), but the MLD signal remains even with respect to the external magnetic field. The amplitude α_{\max} of the MLD as a function of the orientation of incident polarization is plotted in Fig. 2(a). This azimuthal dependence on the incoming polarization allows one to separate the contribution to the MLD related to the spontaneous magnetization from those induced by the external magnetic field. Indeed, when the incoming polarization is parallel to one of the $[100]$ or $[010]$ crystallographic directions, the spontaneous magnetization should have no influence on the magneto-optical response (provided the applied magnetic field is not too strong). For

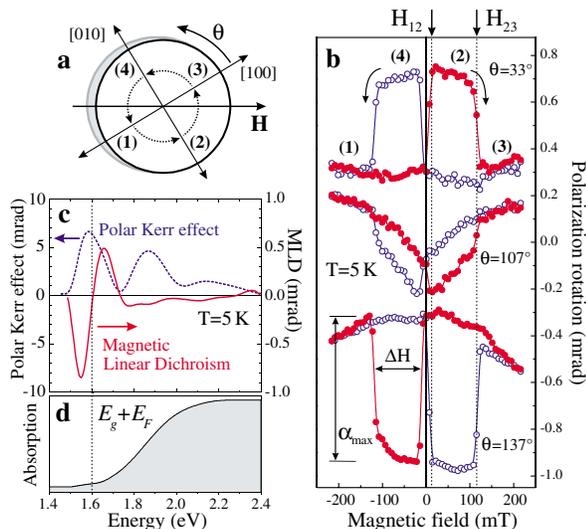


FIG. 1 (color online). (a) $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$ sample orientation with respect to the applied magnetic field and the four-step magnetization reversal process as consecutive 90° jumps (shown by dotted arrows) between the four easy directions (1)–(4). (b) Field dependencies of the magnetic linear dichroism for different angles θ between the incident polarization and the $[100]$ crystallographic direction, measured at a wavelength of $\lambda = 815$ nm. Numbers (1) to (4) correspond to the magnetization directions indicated in (a). H_{12} and H_{23} are the magnetic field values required for making jumps (1) \rightarrow (2) and (2) \rightarrow (3), respectively. (c) Spectra of polar magneto-optical Kerr effect and magnetic linear dichroism (at $\theta = 135^\circ$); (d) absorption spectrum. Note that in panels (c) and (d) Fabry-Perot oscillations in the signal due to the finite buffer thickness have been removed numerically using a bandpass filter.

small angles between the incoming polarization and one of the [100] or [010] crystallographic directions, the field dependence is mostly determined by the contributions proportional to $(\gamma H)^2$ and $\gamma M_{sp} H$ [see curve in the middle of Fig. 1(b)]. However, for angles equal to 45° or 135° one observes only the contribution proportional to M_{sp}^2 . These orientations therefore yield a direct probe of the electronic excitations related to ferromagnetism.

The M-shaped hysteresis loop is observed only in the thermal range below $T_{an} \approx 20$ K, as the fourfold magnetic anisotropy vanishes above that temperature [35]. The temperature increase from 4.2 K up to 20 K therefore results in a rapid decrease of the coercivity of the M-shaped hysteresis, while the magnetization (and therefore also the amplitude of the MLD) stays almost unchanged in this temperature range [see Fig. 2(b)], as the Curie temperature of $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$ is much higher, $T_C = 50$ K. Both critical temperatures T_{an} and T_C depend on the concentration of Mn ions; however, the ratio $T_C/T_{an} \approx 2$ appeared to be fixed [35].

We turn now to the intriguing spectral dependence of the intensity of the MLD signal which is shown in Fig. 1(c), together with the energy dependence of the amplitude of the magneto-optical Kerr effect (all data are taken at 5 K). In the spectral range studied the amplitude of the Kerr effect exhibits two peaks at 1.6 and 1.9 eV, respectively. This energy dependence of the Kerr effect is very similar to the MCD absorption data reported previously [27]. In this paper, it was shown that the low-energy MCD peak around 1.6 eV shifts with the Fermi energy E_F when the hole density is varied [27]. This implies that the 1.6 eV peak is related to interband transitions from valence-band states at the Fermi level to the conduction band. This conclusion is in good agreement with the absorption spectrum at 5 K [see Fig. 1(d)]. Because of the Moss-Burstein shift, the crystal absorbs light only for photon energies $\hbar\omega_0 = E_g + E_F = 1.6$ eV and above, where E_g is the band gap of (Ga, Mn)As. The peak at 1.9 eV in the Kerr spectrum is independent of the location of the Fermi level [27]. We tentatively assign this peak to interband transitions originating deeply within the valence band, where the local density of states exhibits a peak due to hybridization with Mn impurities [7]. The existence of such a feature below the Fermi level is in agreement with several previous experiments [5,6].

The shape of the MLD spectrum is expected to be different from the Kerr effect and MCD spectra. According to Ref. [36], we have for the amplitudes A of MLD and MCD that $A_{MLD} \propto \frac{\partial}{\partial(\hbar\omega)} A_{MCD}$, which appears well obeyed by our experimental findings. A very remarkable observation, however, is the sheer amplitude of the MLD signal: at a photon energy 1.58 eV it amounts to 0.85 mrad. This value is a factor of 4.25×10^2 larger than the MLD observed earlier (0.002 mrad) in (Ga, Mn)As at a—very different—photon energy of 2.28 eV [30]. Note that the MCD [27] and Kerr effect spectra do not reveal such a single sharp peak.

Our observations imply that transitions from states in the vicinity of the Fermi level are almost exclusively responsible for the giant MLD signal in (Ga, Mn)As.

Moreover, it should be mentioned that the MLD in (Ga, Mn)As is also unusually strong in comparison with II–VI diluted magnetic semiconductors such as $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$. One can see from a simple estimate based on previous experiments [31] that a 300 nm film of $\text{Cd}_{0.98}\text{Mn}_{0.02}\text{Te}$ saturated in a strong magnetic field can exhibit an MLD of about 0.01 mrad, but only in the proximity of the exciton resonance, where absorption is strong and all magneto-optical effects are significantly enhanced. In order to understand the very strong MLD signal in (Ga, Mn)As at 1.58 eV, where absorption is relatively weak, one has to consider the origin of this magneto-optical effect in (Ga, Mn)As.

We suggest that the MLD is enhanced when a photo-hole induces an interaction between *two distant* Mn ions. Below we show that this type of interaction is proportional to M^2 . An obvious but important consequence of such a mechanism is that the MLD spectrum will indicate those states in the valence band that are most strongly involved in the Mn-Mn coupling. According to several calculations derived from the Zener model [17,19,23], these states should be in the vicinity of the Fermi level.

For direct band gap semiconductors in the spectral range of the interband transitions, the magneto-optical effects are caused by spin splitting of electron (hole) states in the conduction (valence) bands, $\Delta_{e(h)}$. Therefore, one can write the difference in the absorption indices of Eq. (1) in a power series:

$$\Delta k \propto a\Delta_{e(h)} + b\Delta_{e(h)}^2 + \dots \quad (2)$$

Reference [31] shows that in the paramagnetic phase of DMS, where $\Delta_{e(h)} \propto M$, the coefficient a is equal to zero. This is why MLD is normally negligible compared to first-order magneto-optical effects, such as MCD or the Kerr effect, for which $a \neq 0$. However, for (Ga, Mn)As, where one has carrier-mediated ferromagnetism, the standard

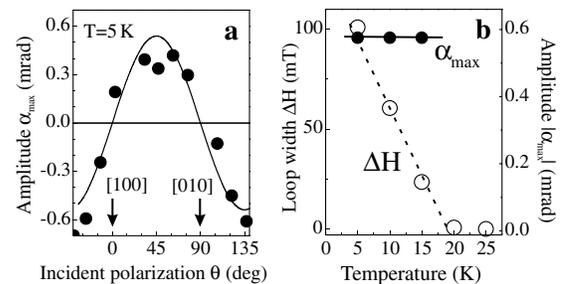


FIG. 2. (a) The amplitude α_{\max} of the linear magnetic dichroism as a function of the angles between the incident polarization and the [100] crystallographic direction in (Ga, Mn)As. (b) Coercivity ΔH and α_{\max} of the M-shaped hysteresis loop as a function of temperature, for an orientation of the incoming polarization of 135° . Lines are guides to the eye.

assumption that the interaction J between the carriers and Mn ions is proportional to the magnetization is expected to be invalid [17,27]. This may lead to a nonzero coefficient a for the linear magnetic dichroism in Eq. (2) and, as a consequence, to a drastic rise of the MLD signal.

In general, the exchange interaction J , to which the splitting $\Delta_{e(h)}$ is proportional, can be written as an expansion:

$$J = \sum A_{ki} s_k S_i + \sum B_{klij} s_k s_l S_i S_j + \dots \quad (3)$$

Here, s is the spin of the itinerant electron (hole), and S_i is the localized magnetic moment provided by the Mn ion at the i th site. The first term in this expansion corresponds to the $sp-d$ exchange interaction, which has been intensively studied for the case of paramagnetic II–VI DMS [1]. Obviously, such a paramagnetic interaction (J_{para}) is proportional to the magnetization.

It is generally accepted that the coupling between distant localized magnetic moments in (Ga, Mn)As is provided by holes. An i th localized magnetic moment induces a spin polarization of an itinerant hole which in turn affects the spin state of another j th localized magnetic moment. Phenomenologically such a ferromagnetic interaction (J_{ferro}) can be described by the second term in Eq. (3), proportional to $s_k^2 S_i S_j$ [37]. This term is quadratic with respect to the magnetization. Thus for (Ga, Mn)As Eq. (3) can be rewritten in a different form:

$$J = J_{\text{para}} + J_{\text{ferro}}, \quad J_{\text{para}} \propto M, \quad \text{and} \quad J_{\text{ferro}} \propto M^2. \quad (4)$$

Because of (4), the first term of Eq. (2) will contain a part proportional to M^2 , contributing to the MLD signal. Such a first-order contribution (but quadratic in the magnetization) will lead to a strong MLD signal in the specific spectral range where photons excite the electronic states that are responsible for the ferromagnetism in these structures. Therefore the giant MLD signal and its predominant sensitivity to these specific electronic states can be explained if the holes in the vicinity of the Fermi level are responsible for the ferromagnetism in (Ga, Mn)As, as is, indeed, expected to be the case. Conversely, this implies that MLD spectroscopy is a very powerful tool to study the valence-band states responsible for ferromagnetism in ferromagnetic semiconductors. In addition, a ratio $J_{\text{para}}/J_{\text{ferro}} \sim 5$ is estimated from the spectral amplitudes of the MLD and the Kerr effect.

In summary, we have found a giant magnetic linear dichroism in (Ga, Mn)As. We show that, in contrast to magnetic circular dichroism, the magnetic linear dichroism is predominantly sensitive to the electronic states responsible for interaction between the spins of magnetic ions. All these observations clearly show that the MLD is a powerful tool for the study of the ferromagnetism related

electronic excitations in (III, Mn)V alloys and similar novel ferromagnetic semiconductor alloys.

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