

## Picosecond Dynamics of the Photoinduced Spin Polarization in Epitaxial (Ga,Mn)As Films

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Static and time-resolved magneto-optical spectra of the ferromagnetic semiconductor (Ga,Mn)As show that a pulsed photoexcitation with a fluence of  $10 \mu\text{J}/\text{cm}^2$  is equivalent to the application of an external magnetic field of about 1 mT, which relaxes with a decay time of 30 ps. This relaxation is attributed to the spin relaxation of electrons in the conduction band and is found to be not affected by interactions with Mn ions.

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The successful growth of the ferromagnetic semiconductor (Ga,Mn)As [1], currently reaching Curie temperatures up to 140 K [2], opens new prospects for the development of novel spintronic devices [3], though at present the fundamental magnetic properties of (Ga,Mn)As are still the subject of intense research [4–14]. The realization of ferromagnetism in a semiconductor results in many unique phenomena which have no analogy in the “standard” physics of magnetism. For instance, the hole-mediated ferromagnetic exchange that occurs in this semiconductor opens the possibility to change the magnetic properties of (Ga,Mn)As by modulation of the hole concentration [14], and a huge photoinduced magnetization of about 15% of the saturation magnetization in ferromagnetic (Ga,Mn)As films was reported recently [13]. The latter, in principle, opens the way to ultrafast manipulation of the magnetic state of (Ga,Mn)As. As was recently demonstrated for (In,Mn)As there are mechanisms of ultrafast nonthermal demagnetization of ferromagnetic semiconductors [15], but up to now no time-resolved analysis of the optically induced magnetization was performed.

In this Letter, we report our observation that the photoexcited nonequilibrium spin polarization in (Ga,Mn)As relaxes with a decay time of 30 ps, which we find to be related to the spin relaxation of electrons in the conduction band. Our observations demonstrate a weak interaction between the spins of the electrons in the conduction band and the spins of the Mn ions, which may have consequences for the application of (Ga,Mn)As in spintronic devices. The spectral characteristics of the Kerr effect induced by an external magnetic field and via photoexcitation are observed to be very similar. Based on this finding we have estimated the equivalent magnetic field that corresponds to the photoexcitation to be about 1 mT. This value is at least 1 order of magnitude less

than the photoinduced magnetization in (Ga,Mn)As reported earlier [13].

The  $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$  sample was grown using a low-temperature molecular beam epitaxy technique described elsewhere [16]. An initial 125 nm GaAs layer and a  $3 \mu\text{m}$   $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$  buffer were deposited on a (001) GaAs substrate at a temperature of  $620^\circ\text{C}$ . These were followed by 10 ML of low-temperature GaAs grown at  $270^\circ\text{C}$ . Subsequently, 350 nm (Ga,Mn)As was deposited, started by opening the shutter of the manganese cell. A reference sample (referred to as LT-GaAs in the text below) was fabricated in exactly the same manner as the (Ga,Mn)As sample, but keeping the manganese shutter closed. The Mn content ( $2\% \pm 1\%$ ) was obtained from a determination of the lattice constant using high resolution x-ray diffraction [16]. The magnetic properties of the sample were characterized by a SQUID magnetometer, revealing ferromagnetic behavior with in-plane easy axes ([100] and [010]) and a Curie temperature of about 50 K. The sample was not annealed.

The magneto-optical Kerr effect (MOKE), that yields a change of the optical polarization of light reflected off a magnetic material, can be used in a pump and probe configuration to monitor the magnetization dynamics. MOKE arises whenever there is a difference between the optical coefficients of a medium for right-handed and left-handed circularly polarized light [17], and is proportional to the magnetization  $M$ :

$$\varepsilon_K + i\theta_K = -\frac{1}{2n(n^2 - 1)}(\chi^{(+)} - \chi^{(-)}), \quad (1)$$

where  $\theta_K$  is the Kerr rotation,  $\varepsilon_K$  is the Kerr ellipticity,  $n$  is the refraction coefficient,  $\chi^{(+)}$  and  $\chi^{(-)}$  are the optical susceptibilities of the material for right-handed and left-handed circularly polarized light, respectively. Since

$[\chi^{(+)} - \chi^{(-)}] \propto M$ , the magneto-optical Kerr effect can be induced not only by the application of an external magnetic field but also by photoexcitation of the medium using circularly polarized light, where a magnetic moment is induced due to the inverse Faraday effect [18,19]. It is this photoinduced MOKE that we employed for our time-resolved studies of the ultrafast magneto-optical properties of (Ga,Mn)As epilayers.

The studies were performed in a “cold-finger” cryostat, mostly at a temperature of 10 K, in the spectral range between 1.49 and 1.66 eV. A few experiments were done at 60 K. The time-resolved measurements were performed in a pump and probe configuration, using pulses from a Ti:sapphire laser with a pulse length of approximately 100 fs and a repetition rate of 76 MHz. The setup is described in detail elsewhere [20]. In brief, the polarization of the circularly polarized pump beam was modulated between left-handed and right-handed helicities, while the probe beam was linearly polarized. Both beams were focused onto the same spot on the sample; the spot diameter was 100  $\mu\text{m}$  for the pump and slightly less for the probe. The power density per pump pulse was around 0.1  $\text{GW}/\text{cm}^2$ , which corresponds to a fluence of 10  $\mu\text{J}/\text{cm}^2$ . The Kerr rotation and ellipticity of the reflected probe beam were measured as a function of the time delay between pump and probe pulses. In order to investigate the static magneto-optical properties of the (Ga,Mn)As film, the MOKE response induced by an external magnetic field was studied as well. In these experiments, an external magnetic field of up to 100 mT, provided by an electromagnet, was applied perpendicular to the plane of the sample. The static measurements were performed at normal incidence of the light, i.e., in a polar configuration [17]. The MOKE as a function of temperature was measured for such small light intensity that both accumulated heat and photoinduced changes of the Curie temperature could be neglected. These data were used later to monitor variations of the magnetic properties and the temperature rise of the photoexcited area. The latter was found to be less than 10 K during the pump-probe measurements.

Experimental data of the dynamics of the photoinduced Kerr rotation of the (Ga,Mn)As film at 10 K are shown in Fig. 1 for zero magnetic field (circles). One observes that the Kerr rotation relaxes with a decay time of about 30 ps.

In order to separate the photoinduced signal from the magnetic layer from that of the other layers in the sample, we repeated the experiment using the reference structure containing a LT-GaAs layer instead of (Ga,Mn)As. The results of these measurements are also included in Fig. 1 (diamonds). Figure 1 shows that the temporal dependencies of the Kerr effect from the structures with and without manganese are quite different. Such substantial difference between the temporal dependencies for these two structures can be related to Mn-induced changes of

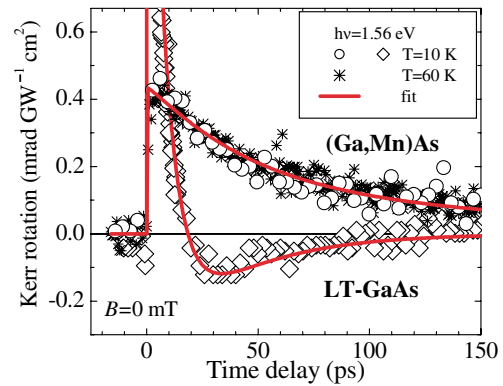


FIG. 1 (color online). Dynamics of the Kerr rotation from the  $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$  film recorded at temperatures of 10 and 60 K (circles and stars), compared with similar data from the reference sample containing a LT-GaAs film measured at 10 K (diamonds). The photon energy was 1.56 eV. The solid lines are fits to the experimental data.

the lattice constant and an increase of defect density, that result in a significant difference between the optical properties of (Ga,Mn)As and LT-GaAs. Note that the relaxation of the magneto-optical signal in both cases is much faster than that observed by us from the (001) GaAs substrate (of about 250 ps). These observations prove that for the structure with a magnetic film the photoinduced Kerr effect is dominated by a signal related intrinsically to the magnetic (Ga,Mn)As layer.

For a more systematic analysis the temporal dependencies measured at different photon energies were fitted as a sum of two exponential decays with characteristic times  $\tau_1 \sim 7$  and  $\tau_2 \sim 30$  ps and different amplitudes  $I_1$  and  $I_2$  [21], respectively, i.e.,

$$I = I_1 \exp(-t/\tau_1) + I_2 \exp(-t/\tau_2). \quad (2)$$

Because of energy redistribution of the photoexcited carriers, the Kerr effect can be very much affected by a change of optical properties, while the magnetization stays unchanged [22]. Therefore, the interpretation of the faster  $\tau_1$  component of the Kerr effect is not trivial and needs a more detailed analysis, as discussed previously [20]. In the present Letter we limit ourselves to a discussion of the  $\tau_2$  component of the Kerr effect, which is attributed to spin relaxation processes.

The spectral behavior of  $I_2$  is shown in Fig. 2(a). The spectra clearly show several oscillations, which do obey the Kramers-Kronig relations. At the energies at which the photoinduced Kerr rotation ( $\theta_K$ ) changes sign, the photoinduced Kerr ellipticity ( $\varepsilon_K$ ) reaches a negative extremum.

The spectral dependencies of the Kerr rotation and the Kerr ellipticity induced by an external magnetic field in the (Ga,Mn)As film are shown in Fig. 2(b). The observed dependencies qualitatively reproduce the spectra of the photoinduced signal. The similarity of the spectra allows

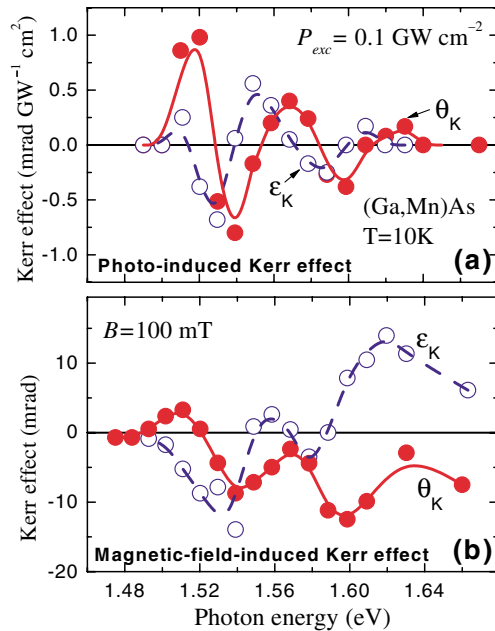


FIG. 2 (color online). (a) Experimental spectral dependencies of the contribution with decay of 30 ps to the photoinduced Kerr rotation (solid circles) and ellipticity (open circles) from the  $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$  sample. (b) Experimental spectral dependencies of the magnetic field induced Kerr rotation (solid circles) and ellipticity (open circles) from the  $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$  sample. Lines are guides to an eye.

us to compare the two effects and to extract an equivalent magnetic field created by the photoexcitation. From a comparison of the magnetic field induced and photoinduced Kerr effects one can obtain that an excitation with a fluence of  $10 \mu\text{J}/\text{cm}^2$  (density  $0.1 \text{ GW}/\text{cm}^2$ ) is equivalent to the application of an external magnetic field of about  $0.6 \pm 0.4 \text{ mT}$ , that is below  $1 \text{ mT}$  [23].

The field dependence of the Kerr effect measured in a static external magnetic field at a photon energy of  $1.60 \text{ eV}$  is presented in Fig. 3. One can distinguish a linear increase of the magneto-optical signal in magnetic fields up to  $35 \text{ mT}$  (with a slope of  $0.05 \text{ mrad}/\text{mT}$ ), followed by a jump and a saturation at  $75 \text{ mT}$ . This linear increase in low magnetic fields and jumps slightly below  $0.1 \text{ T}$  are in agreement with results of previous studies of magnetic properties of  $(\text{Ga},\text{Mn})\text{As}$  [5,10]. From these data one can see that the detected photoinduced signal is about  $0.05 \pm 0.005 \text{ mrad}$  at an excitation power of  $0.1 \text{ GW}/\text{cm}^2$ . It shows that the photoinduced magnetization is just  $0.4\%$  of the saturation magnetization in  $(\text{Ga},\text{Mn})\text{As}$ . Such a value of the photoinduced magnetization is a factor of 40 smaller than the one observed in the experiment with less powerful but continuous optical excitation [13]. This discrepancy could be due to the fact that the phenomenon studied earlier is due to slow processes and cannot be observed in the subnanosecond time domain. Moreover, we should also note that the discrepancy may originate from the fact that in [13] a transport measurement (i.e.,

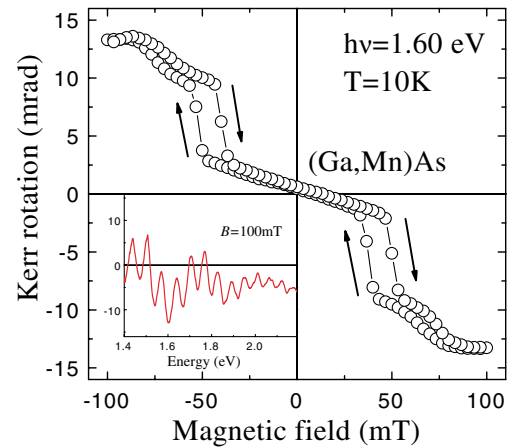


FIG. 3 (color online). Magnetic field induced Kerr rotation of the epitaxial  $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$  film as a function of magnetic field applied perpendicular to the plane of the sample. Inset: spectrum of that measured at  $100 \text{ mT}$ .

the anomalous Hall effect) was used to infer the change in magnetization. However, such transport measurements may be compromised by photosensitivity of the contact resistance.

The photoinduced Kerr effect from the  $(\text{Ga},\text{Mn})\text{As}$  layer can contain contributions from polarized photoexcited carriers and from the alignment of magnetic ions. In order to separate these two contributions we measured the dynamics of the photoinduced Kerr rotation at a temperature of  $60 \text{ K}$  (the stars in Fig. 1). At this temperature the  $(\text{Ga},\text{Mn})\text{As}$  layer is paramagnetic, which should significantly change the temporal behavior of the photoinduced MOKE associated with Mn ions. It is clear from Fig. 1 that the temperature increase does not result in any significant changes in the photoinduced Kerr effect and the observed difference is within the limits of the experimental error. This demonstrates that the observed signal is not related to the magnetization of Mn ions and thus must be attributed to the photoexcited carriers only. Because of the strong spin-orbit interaction as well as the coupling of the quasimomentum and the angular momentum of the holes in III-V compounds, the spin relaxation time of holes in bulk semiconductors of the GaAs type is usually much shorter than that of electrons [24,25] and in the studied thermal range it is expected to be about  $1 \text{ ps}$  [24]. Thus the observed photoinduced magneto-optical signal with the relaxation time of  $30 \text{ ps}$  cannot be associated with holes and must be assigned to the spin-polarization decay of conduction-band electrons.

In order to reveal a possible interaction between the spins of the photoexcited electrons and those of the Mn ions, we repeated the measurements of the photoinduced Kerr effect in an external magnetic field up to  $80 \text{ mT}$ , applied in the plane of the sample along the  $[110]$  axis. This field saturates the in-plane magnetization of the sample. In such a geometry, any interaction of Mn ions

with spin polarized photoexcited carriers should result in the excitation of a precession of manganese spins around the field and vice versa, of the electron spins around manganese moments [26]. However, we did not observe any oscillations nor any significant modification of the relaxation of the photoinduced signal. This implies that an interaction of the Mn spins with the spins of electrons in the conduction band is rather weak [27].

This finding is in good agreement with the results of electronic structure calculations of (Ga,Mn)As, which shows that the 3D states of Mn are strongly hybridized with the valence band, while their influence on the conduction band is much weaker [28]. Therefore, spintronic devices based on (GaMn)As can only operate using hole currents. Regarding the lower mobility of holes in comparison with that of electrons, this necessary condition will limit the operation speed of (Ga,Mn)As-based devices, unless sophisticated design solutions are applied [29–31].

In summary, the photoinduced spin polarization in epitaxial (Ga,Mn)As films was found to be equivalent to the application of an external magnetic field of about 1 mT for an optical excitation with a fluence of  $10 \mu\text{J}/\text{cm}^2$ . This photoinduced magnetization is 1 order of magnitude less than the one reported earlier, and is found to relax with a decay time of 30 ps, which is attributed to the spin relaxation of electrons in the conduction band. Raising the sample temperature above the Curie point, as well as application of a large in-plane magnetic field does not result in any significant modification of this relaxation. From this we conclude that the spin relaxation of electrons is not much affected by interactions with Mn ions. This conclusion will have important consequences for possible application of (Ga,Mn)As in spin-electronic devices.

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- [1] H. Ohno *et al.*, Appl. Phys. Lett. **69**, 363 (1996).
  - [2] K.W. Edmonds *et al.*, Appl. Phys. Lett. **81**, 4991 (2002).
  - [3] Special issue on semiconductor spintronics [Semicond. Sci. Technol. **17**, 275 (2002)].
  - [4] T. Hayashi, M. Tanaka, K. Seto, T. Nishinaga, and K. Ando, Appl. Phys. Lett. **71**, 1825 (1997).
  - [5] K. Ando, T. Hayashi, M. Tanaka, and A. Twardowski, J. Appl. Phys. **83**, 6548 (1998).
  - [6] B. Beschoten *et al.*, Phys. Rev. Lett. **83**, 3073 (1999).
  - [7] T. Dietl, H. Ohno, and F. Matsukura, Phys. Rev. B **63**, 195205 (2001).
  - [8] D. Hrabovsky *et al.*, Appl. Phys. Lett. **81**, 2806 (2002).

- [9] X. Liu, Y. Sasaki, and J.K. Furdyna, Phys. Rev. B **67**, 205204 (2003).
- [10] M. Sawicki *et al.*, cond-mat/0212511.
- [11] U. Welp, V.K. Vlasko-Vlasov, X. Liu, J.K. Furdyna, and T. Wojtowicz, Phys. Rev. Lett. **90**, 167206 (2003).
- [12] H. X. Tang, R. K. Kawakami, D. D. Awschalom, and M. L. Roukes, Phys. Rev. Lett. **90**, 107201 (2003).
- [13] A. Oiwa, Y. Mitsumori, R. Moriya, T. Slupinski, and H. Munekata, Phys. Rev. Lett. **88**, 137202 (2002).
- [14] H. Ohno *et al.*, Nature (London) **408**, 944 (2000).
- [15] J. Wang, G.A. Khodaparast, J. Kono, T. Slupinski, A. Oiwa, and H. Munekata, cond-mat/0305017.
- [16] G.M. Schott, W. Faschinger, and L.W. Molenkamp, Appl. Phys. Lett. **79**, 1807 (2001).
- [17] A.K. Zvezdin and V.A. Kotov, *Modern Magnetooptics and Magneto-optical Materials* (IoP Publishing, Bristol, 1997).
- [18] L.P. Pitaevskii, Sov. Phys. JETP **12**, 1008 (1961).
- [19] L.D. Landau and E.M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, Oxford, 1984).
- [20] A.V. Kimel, V.V. Pavlov, R.V. Pisarev, V.N. Gridnev, F. Bentivegna, and Th. Rasing, Phys. Rev. B **62**, R10610 (2000).
- [21] A.V. Kimel, F. Bentivegna, V.N. Gridnev, V.V. Pavlov, R. V. Pisarev, and Th. Rasing, Phys. Rev. B **63**, 235201 (2001).
- [22] B. Koopmans, M. van Kampen, J.T. Kohlhepp, and W.J.M. de Jonge, Phys. Rev. Lett. **85**, 844 (2000).
- [23] The measurements of the magnetic field induced MOKE between 1.4 and 2.2 eV (see inset of Fig. 3) clearly demonstrate that the oscillatory behavior in Fig. 2 is caused by an optical interference. In spite of this fact, the spectra of the two effects are affected by the interference in a similar way (see Fig. 2) and thus can be compared.
- [24] *Optical Orientation*, edited by F. Meyer and B.P. Zakharchenya (North-Holland, Amsterdam, 1984).
- [25] D.J. Hilton and C.L. Tang, Phys. Rev. Lett. **89**, 146601 (2002).
- [26] J.M. Kikkawa and D.D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998).
- [27] Note that the strong interaction between the spins of Mn and that of holes is responsible for ferromagnetism in (Ga,Mn)As. Therefore the absence of magnetic field dependence of the photoinduced MOKE once again demonstrates that the observed signal cannot be assigned to the photoexcited holes. Otherwise, the strong coupling between the spins of holes and those of Mn ions should result in some modification of the temporal behavior of the MOKE, even if the damping of the homogeneous precession is fast.
- [28] E. Kulatov, H. Nakayama, H. Mariette, H. Ohta, and Yu. A. Uspenskii, Phys. Rev. B **66**, 045203 (2002).
- [29] M. Kohda, Y. Ohno, K. Takamura, F. Matsukura, and H. Ohno, Jpn. J. Appl. Phys. **40**, L1274 (2001).
- [30] E. Johnston-Halperin *et al.*, Phys. Rev. B **65**, 041306 (2002).
- [31] P. van Dorpe *et al.*, Appl. Phys. Lett. (to be published).