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Ultrafast optical control of coercivity in GaMnAs

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Femtosecond optical control of the magnetization and coercive field is demonstrated in GaMnAs using time-resolved magneto-optical Kerr effect techniques. These experiments reveal a near-complete, subpicosecond collapse of the hysteresis loop, consistent with femtosecond demagnetization. On longer time scales (~ 300 ps) an increase in coercivity is observed, attributed to hole-mediated enhancement of the domain wall energy. © 2008 American Institute of Physics. [DOI: 10.1063/1.2963972]

In the III-Mn-V diluted magnetic semiconductors (DMSs), the ferromagnetic interaction between the magnetic impurities is mediated by holes, leading to a unique combination of semiconducting and magnetic properties.^{1,2} As a result, unlike ferromagnetic metals, ferromagnetic order in DMS materials may be controlled using the well-established methods of carrier density control in semiconductors, leading to a host of possibilities for future multifunctional devices.³⁻⁸ Hole-mediated enhancement of the Curie temperature (T_C) has been demonstrated using electrical gates⁹ and continuous-wave optical excitation.¹⁰ More recently, femtosecond optical experiments have revealed ultrafast magnetization quenching, and a transient enhancement of both magnetization and T_C .^{11,12}

The magnetic properties governing magnetization reversal should also be subjected to control through variation of the carrier density. Predictions based on the Zener mean field model^{2,14,15} show that both the exchange constant (or spin stiffness) A and the anisotropy constant K_u are highly sensitive to changes in hole density, indicating the ability to actively control the coercive field (H_c). In InMnAs, gate control of H_c was demonstrated by Chiba *et al.*¹³ and a semipermanent change in H_c was observed by Oiwa *et al.* following excitation by a continuous-wave diode laser.¹⁶ The combination of these effects with ultrafast optical demagnetization would lead to a versatile platform for ultrafast magneto-optical data storage.^{8,17} Studies that extend the quasiequilibrium experiments in Refs. 13 and 16 to the subnanosecond temporal regime are needed.

We present here results of femtosecond optical studies of the magnetization reversal process in GaMnAs. Unlike earlier ultrafast studies in this system,¹² tensile strain leads to perpendicular uniaxial anisotropy in the structure studied in this work, facilitating a direct measurement of the magnetic hysteresis loop as a function of time delay following femtosecond optical excitation. A similar approach was used in studies on InMnAs by Wang *et al.*, who observed a transient reduction of H_c that persisted for ~ 2 ps.¹⁸ The experiments reported here on GaMnAs were carried out in a much higher excitation regime, revealing dramatic optically induced changes to the magnetic response. Our results indicate a nearly complete collapse of the hysteresis loop on the time scale of the optical pulse, reflecting a strong ultrafast reduc-

tion in ferromagnetic order. The magnetic hysteresis loop is observed to broaden on longer time scales (~ 300 ps), indicating an optically induced increase in H_c . We attribute this effect to hole-mediated enhancement of the domain wall energy. These experiments provide insight into the ferromagnetic properties of DMS materials in the nonequilibrium regime. Our demonstration of ultrafast control of coercivity is promising for possible applications of DMS materials in future multi-functional devices.

The sample studied in the present work was grown by low temperature molecular beam epitaxy, and consists of a 270 nm thick layer of $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ ($x \sim 0.03$) grown on a 3.5 μm thick ZnSe buffer layer on a semi-insulating GaAs substrate. The ZnSe buffer layer induces tensile strain in the GaMnAs layer, causing the easy axis of magnetization to lie along the growth direction.^{1,2,14,19} Results of continuous-wave magneto-optical Kerr effect measurements are shown in Fig. 1.²⁰ The observation of square hysteresis loops indicates strong perpendicular uniaxial anisotropy in this structure. The background hole concentration estimated from magneto-transport data is $\sim 1 \times 10^{20} \text{ cm}^{-3}$. The Curie temperature of the structure studied here is approximately 45 K.

The magnetization dynamics were measured using pump-probe magneto-optical Kerr effect experiments at 10 K. The optical source used produces 60 fs, 800 nm pulses at 250 kHz. A white light continuum was generated using a small portion of the laser output and used as a tunable probe beam. The 800 nm output of the laser system was used directly for the pump beam. The pump pulses excite low en-

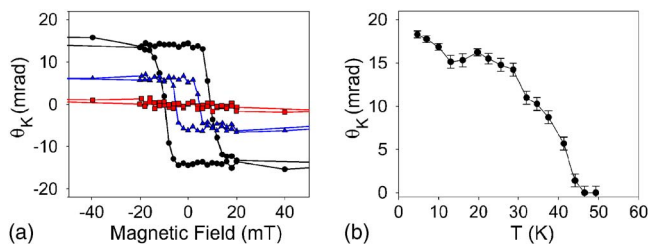


FIG. 1. (Color online) Temperature dependent Kerr rotation measurements on GaMnAs taken with the pump beam blocked. The magnetic field is applied along the growth direction. (a) Hysteresis loops for 10 K (circles), 37 K (triangles), and 46 K (squares); (b) Remanent magnetization vs sample temperature, indicating $T_C \sim 45$ K.

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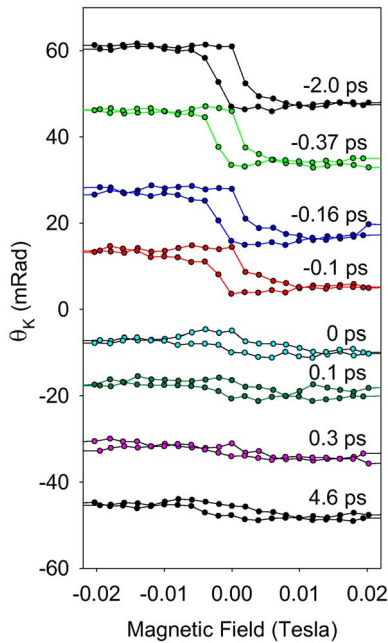


FIG. 2. (Color online) Results of time-resolved Kerr rotation studies on GaMnAs at 10 K, showing the dependence of the hysteresis loop on probe time delay. The loops have been offset by equal increments for clarity. Negative delay values correspond to the arrival of the probe pulse before the pump pulse.

ergy electron hole pairs in the GaMnAs epilayer. The pump fluence was $\sim 15 \text{ mJ/cm}^2$. Using the calculated absorption spectrum for *p*-doped GaAs,² the optically-injected hole density is estimated to be $2 \times 10^{20} \text{ cm}^{-3}$. This is an upper estimate only as saturation effects are expected to be significant for these pumping conditions. The Kerr rotation signal was measured at 610 nm using a standard polarization bridge. Differential reflectivity experiments were also performed at a probe wavelength of 715 nm, yielding information about the carrier trapping and recombination dynamics.

The pump-induced change in the magnetization reversal process is shown in Fig. 2. The pump has two notable effects on the hysteresis loop: (i) the amplitude of the remanent magnetization is strongly reduced and (ii) the abrupt magnetization reversal at the coercive field is replaced by a gradual transition for positive probe pulse delays. The hysteresis loop for positive delays resembles a paramagnetic response, indicating a strong pump-induced reduction in magnetic order. The onset of these effects is instantaneous within our time resolution. The results in Fig. 2 are similar to earlier experiments in CoPt₃ alloy films, in which a full collapse of the hysteresis loop was observed within 120 fs.²¹

The pump-induced change in magnetization ($\Delta\theta_K$) is shown in Fig. 3(a) for a short range of time delays following the arrival of the pump pulse. A contribution to the signal exists near zero time delay that does not reverse sign with the reversal of the magnetic field. This signal represents a transient downward shift of the entire hysteresis loop. The average of the $\Delta\theta_K$ signals for $\pm 180 \text{ mT}$ is shown in Fig. 3(b) (circles). The zero delay peak follows the pulse cross correlation [dashed curve in Fig. 3(b)] and is therefore likely due to two photon absorption. The slower component in Fig. 3(b) decays exponentially with a time constant of 1 ps, and is attributed to fast electron trapping due to ionized antisite defects introduced during low temperature growth.^{22–24} Aside from the picosecond transient shift of the hysteresis

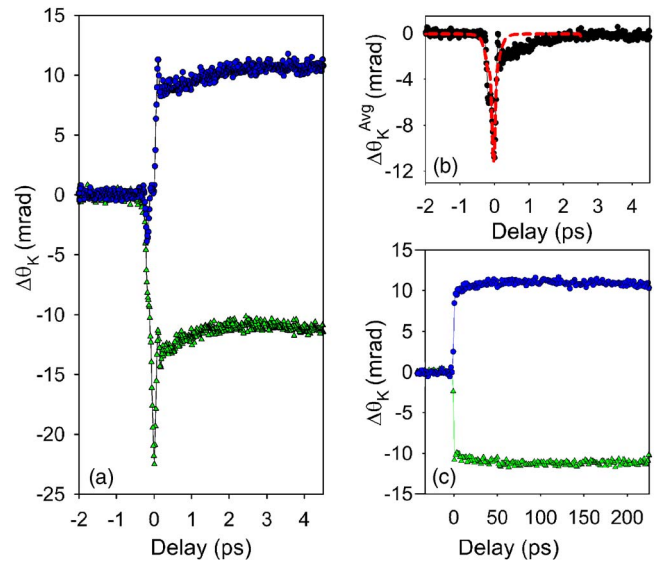


FIG. 3. (Color online) (a) The pump-induced change in the probe Kerr rotation ($\Delta\theta_K$) vs time delay. The magnetic field is $+180 \text{ mT}$ (circles) or -180 mT (triangles). (b) The average of the two scans in (a) (circles) is plotted together with the cross correlation between the pump and probe beams (dashed curve). (c) Same data as in (a) for a larger range of probe delay. The sample temperature is 10 K.

loop, the $\Delta\theta_K$ signal is symmetric with respect to reversal of the magnetic field direction, and disappears above T_C . The observed $\Delta\theta_K$ signal is therefore dominated by pump-induced changes in magnetic order. This was further verified by comparing $\Delta\theta_K$ and the pump-induced change in Kerr ellipticity ($\Delta\eta_K$), which exhibit identical temporal dynamics. Beyond the region of pulse overlap, $\Delta\theta_K$ is $\sim 70\%$ of the loop height before zero delay. The ferromagnetic to paramagnetic phase transition is therefore not complete, consistent with the appearance of a weak residual hysteresis in the loops for positive time delays in Fig. 2. $\Delta\theta_K$ decays (i.e., the magnetization recovers) due to the external magnetic field on a much longer time scale ($\sim 5 \text{ ns}$). The evolution of $\Delta\theta_K$ is similar to that observed in InMnAs,¹¹ although no details of the magnetization reversal process were reported. The evolution of the hysteresis loop on longer time scales is shown in Fig. 4. The remanent magnetization has partially recovered after 2 ns delay [Fig. 4(a)], consistent with the time scale for decay of $\Delta\theta_K$. The coercive field *increases* with time delay, as shown in Fig. 4(b). For delays longer than a few hundred picoseconds, H_c is enhanced by approximately three times relative to the value before the arrival of the pump pulse.

The carrier density injected by the pump pulse is comparable to the background hole density. The influence of

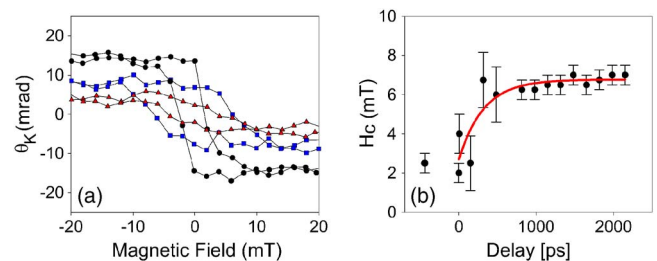


FIG. 4. (Color online) (a) Hysteresis loops for probe delays of -2 ps (circles), $+4 \text{ ps}$ (triangles), and $+2 \text{ ns}$ (squares). (b) Coercive field vs probe delay. The rise time obtained from the exponential fit (solid curve) is 320 ps.

these excess carriers on the magnetic properties is therefore expected to be significant. The optically injected holes lead initially to a strong demagnetization through the reverse Overhauser effect, which leads to a net transfer of spin polarization from the Mn ions to the hole population.^{11,12} After the first few picoseconds, the holes begin to thermalize and relax into the spin split bands. These excess holes modify the underlying parameters governing ferromagnetism, such as the Mn–Mn exchange coupling, the transition temperature, and the anisotropy energy. Fitting the data in Fig. 4(b) to an exponential rise (solid curve) indicates a build-up time for the coercivity enhancement of 320 ps, attributed to the hole thermalization process. The holes recombine on a much longer time scale.^{12,22} Our differential reflectivity measurements indicate a recombination time of 6.5 ns for this structure. As discussed below, the enhancement of coercivity for positive probe pulse delays is attributed to a transient hole-mediated increase in the domain wall energy.

The magnetization reversal process in GaMnAs films with perpendicular uniaxial anisotropy occurs via the formation and propagation of Bloch domain walls.^{25,26} The coercive field is most likely limited by domain wall pinning at defects. Using a simple micromagnetic description,²⁷ the coercive field $H_c = c(\sigma_{dw}/M_s)$, where $\sigma_{dw} = 4(AK_u)^{1/2}$ is the domain wall energy, M_s is the saturation magnetization, and c is a proportionality constant that describes the nature of the pinning defects (i.e., the defect dimension and the fractional fluctuation in the domain wall energy). The effects of the pump beam vanish reversibly when it is blocked, indicating that the optical excitation process has no effect on the defect distribution in the sample. For GaMnAs layers under tensile strain and our conditions of background hole density and pump fluence, A and K_u are both predicted to increase strongly with increasing hole density,^{2,15} leading to an increase in σ_{dw} (and thus H_c) for larger hole densities. The observation of larger coercivities for positive time delays [Fig. 4(b)] is therefore attributed to hole-mediated enhancement of the domain wall energy. The calculations in Refs. 2 and 15 correspond to the properties of ferromagnetism in equilibrium, in contrast to the strongly nonequilibrium conditions of the present experiments. Nevertheless, the general trend predicted is consistent with our observation of enhanced coercivity for positive time delays. No decay of the coercivity enhancement is observed within the accessible range of delays. This is reasonable considering the long carrier recombination time in the structure studied here.

We note that the coercivity for negative time delays is reduced from the value when the pump beam is blocked. Due to the relatively large 4 μ s time interval between pulses, this reduction must be due to a slow process. The coercivity was reduced to a greater extent than the remanent magnetization, although the loop remains square, indicating that the perpendicular anisotropy is maintained. Simple heat accumulation is not sufficient to account for this reduction. Nevertheless, the observation of a transient increase in H_c following optical excitation supports our interpretation based on hole-mediated enhancement.

Correlated Mn precession was recently observed in GaMnAs,^{28–30} attributed to a hole-mediated change in the easy axis direction. In the work reported here, only the magnitude of K_u is affected by the optically injected holes due to the strong perpendicular uniaxial anisotropy. Consequently,

no precessional motion of the Mn is expected for our structure.

In summary, femtosecond optical control of the magnetization and coercive field in GaMnAs was demonstrated using time-resolved magneto-optical Kerr effect techniques. A pulse-width-limited collapse of the hysteresis loop was observed, followed by enhancement of the coercive field on longer time scales. We attribute the observed increase in coercivity to hole-mediated enhancement of the domain wall energy. These results appear promising for the development of future multifunctional devices using DMS materials.

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