Ultrafast Quenching of Ferromagnetism in InMnAs Induced by Intense Laser Irradiation

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Time-resolved magneto-optical Kerr spectroscopy of ferromagnetic InMnAs reveals two distinct *demagnetization* processes—fast (<1 ps) and slow (~100 ps). Both components diminish with increasing temperature and are absent above the Curie temperature. The fast component rapidly grows with pump power and saturates at high fluences (>10 mJ/cm^2); the saturation value indicates a *complete quenching* of ferromagnetism on a subpicosecond time scale. We attribute this fast dynamics to spin heating through *p-d* exchange interaction between photocarriers and Mn ions, while the ~100 ps component is interpreted as spin-lattice relaxation.

There is much current interest in dynamical processes in *magnetically ordered* systems, both from scientific and technological viewpoints [1]. Pumping a magnetic system with ultrashort laser pulses can strongly alter the equilibrium among the constituents (carriers, spins, and the lattice), triggering a variety of dynamical processes. Studying these processes can provide estimates for the time scales and/or strengths of the various interactions. Both metallic and insulating magnets have been studied, exhibiting an array of new phenomena. In particular, the discovery of *ultrafast demagnetization* [2] suggested an ultrafast scheme for magneto-optical data writing. At the same time, exactly how a laser pulse can effectively change the magnetic moment is a matter of debate [3–6].

Despite the large number of studies performed to date, the microscopic understanding of effective energy transfer channels among the subsystems is still elusive. This is partially due to the fact that, in the case of metallic ferromagnets, the distinction between the "carriers" and the "spins" is subtle, as the itinerant electrons contribute both to transport and magnetism. On the other hand, antiferromagnetic insulators have shown much slower dynamics, typically on the order of hundreds of ps [7], although femtosecond demagnetization has been predicted [8], and ultrafast control of magnetism using off-resonance excitation has been shown experimentally [9]. Carrier-mediated ferromagnetism in (III,Mn)V semiconductors provides an interesting alternative for this type of study. Unlike in ferromagnetic metals, there is a clear distinction between mobile carriers (holes) and localized spins (Mn ions), and ferromagnetic order is realized through their strong coupling (*p-d* exchange interaction). This coupling in turn makes the magnetic order sensitive to carrier density changes via external perturbations [10–13].

In this Letter, we describe time-dependent magneto-optical studies of laser-induced demagnetization dynamics in ferromagnetic semiconductor InMnAs. We observed a rapid (<1 ps) drop of magnetization along with a slower demagnetization process on the order of ~100 ps. Both demagnetization components show strong temperature and pump power dependence. At high pump fluences and under external magnetic fields, a complete quenching of the ferromagnetic phase occurs, which appears as a saturation of the photoinduced Kerr rotation with pump fluence. We attribute the observed fast and slow processes to the manifestations of hole-localized spin and spin-lattice interactions, respectively.

The main sample studied was an In$_{0.87}$Mn$_{0.13}$As(25 nm)/GaSb(820 nm) heterostructure with Curie temperature ~60 K, grown by low-temperature molecular beam epitaxy on a semi-insulating GaAs (100) substrate [14]. Taking into account the electrical and magnetic influences of interstitial Mn ions which are supposed to be double donors and couple antiferromagnetically with substitutional Mn ions [15–17], the hole density p was extracted analytically from the saturation magnetization data of the ferromagnetic component. In the present sample, $p \simeq 4 \times 10^{20}$ cm^{-3}, and the density of Mn spins participating in ferromagnetic order is $\simeq10^{21}$ cm^{-3}.

The two-color time-resolved magneto-optical Kerr effect (MOKE) spectroscopy setup used consisted of an optical parametric amplifier (OPA) pumped by a Ti:Sapphire-based regenerative amplifier (Model CPA-2010, Clark-MXR, Inc.). The OPA pump beam, which was linearly polarized and tuned to 2 μm, excited transient carriers near the band edge of InMnAs, and a very small fraction ($\sim10^{-5}$) of the regenerative amplifier beam (775 nm) was used as a probe; the high photon energy of the probe ensured to diminish the "dichroic bleaching" effects [3] due to the pump excited carriers.

Typical data showing the general temporal profile of the photoinduced Kerr angle change, $\Delta\theta_K$, are shown in Fig. 1, for the first 3 ps [in panel (a)] and the entire time range [in panel (b)]. The sign of $\Delta\theta_K/\theta_K$ is always negative, indi-
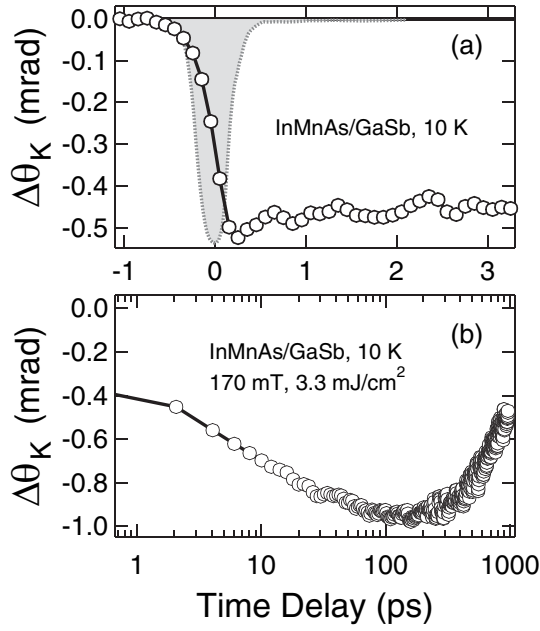
FIG. 1.   (a) The first 3 ps of demagnetization dynamics in InMnAs/GaSb. Also shown is the cross correlation between the pump and probe pulses. (b) Demagnetization dynamics covering the entire time range of the experiment (up to ~1 ns). There is a slow demagnetization process, which follows the fast component shown in (a) and completes only after ~100 ps.
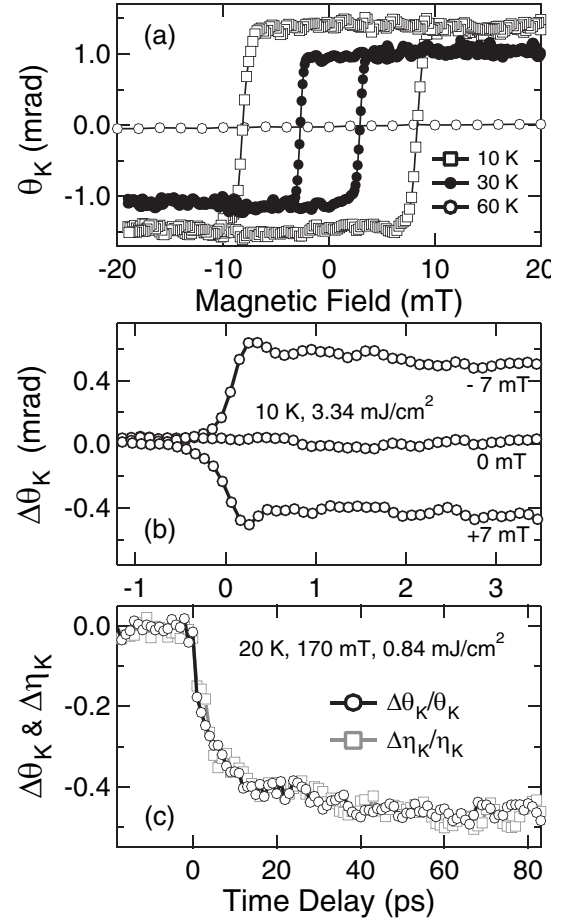

FIG. 2.   (a) Steady-state MOKE angle versus magnetic field, taken with a 775 nm laser diode. (b) Magnetic field dependence of ultrafast demagnetization dynamics. The sign of the MOKE angle change depends on the direction of the applied magnetic field in a symmetric manner, as expected. (c) Comparison between MOKE (θ_K) and MCD (η_K) dynamics, showing no time lag between them.

cating transient *demagnetization*. Distinct temporal regimes can be identified: an initial (<1 ps) reduction in magnetization is followed by a slow and gradual decrease (up to ~100 ps), a plateau region (up to ~500 ps), and, finally, an increase (i.e., recovery) toward the equilibrium value. Also shown in Fig. 1 is the cross-correlation trace between the pump and probe pulses, showing that the initial ultrafast demagnetization is occurring even faster than our time resolution (~220 fs).

Figure 2(a) shows cw MOKE data taken with a 775 nm laser diode. The MOKE angle is ~1.4 mrad and the coercivity is ~7.5 mT at 10 K. MOKE dynamics in the first 3 ps is shown in Fig. 2(b) for different magnetic fields: +7, 0, and −7 mT. The sign of $\Delta\theta_K$ changes when the direction of the field is reversed, as expected for demagnetization. In order to eliminate the possibility that the induced MOKE changes are due to purely optical effects, we also monitored both MOKE and reflection magnetic circular dichroism (MCD) at the same time. The exact coincidence between MOKE and MCD, shown in Fig. 2(c), ensures that we are probing magnetic properties (see, e.g., [3]).

Photoinduced Kerr rotation dynamics, normalized by the MOKE angle before the arrival of the pump, is plotted for different pump fluences in Fig. 3(a). At low pump fluences, the fast (<1 ps) and slow (~100 ps) demagnetization components coexist. However, as the fluence increases, the fast component progressively becomes more dominant. Around ~10 mJ/cm^2, there is no slow demag-

netization process any more—a sharp initial drop is followed by a completely flat region. This "step-function"-like response remains stable both in shape and magnitude against further increases in pump fluence to 13.4 mJ/cm^2; i.e., the demagnetization saturates. The saturation value is 1, implying that the change in magnetization is 100% [see also Fig. 3(b)]. These results suggest that a *complete quenching* of the ferromagnetic order is occurring on the order of several hundred fs.

To substantiate the above claim, we performed temperature dependent measurements, and the results are shown in Fig. 4(a). Here, a strong temperature dependence of both the fast and slow components is seen. $\Delta\theta_K$ decreases drastically as the temperature approaches the critical temperature T_c(~60 K), and $\Delta\theta_K$ is *absent above the T_c*. These facts further corroborate that the transient Kerr rotation here measures the magnetization change of the ferromagnetic state [Fig. 4(b)].
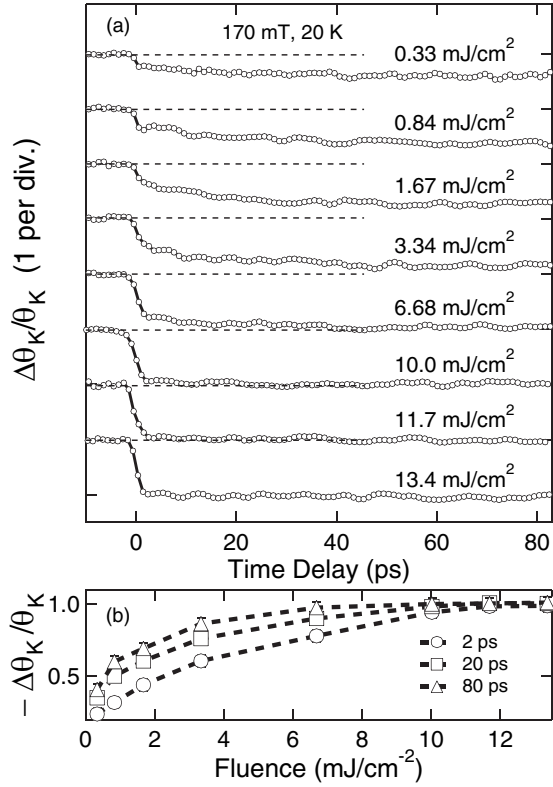
FIG. 3. Normalized photoinduced MOKE angle change (a) versus time for different pump fluences and (b) versus pump fluence for different time delays. At high fluences, the signal saturates to ~1, suggesting a complete quenching of ferromagnetic order.
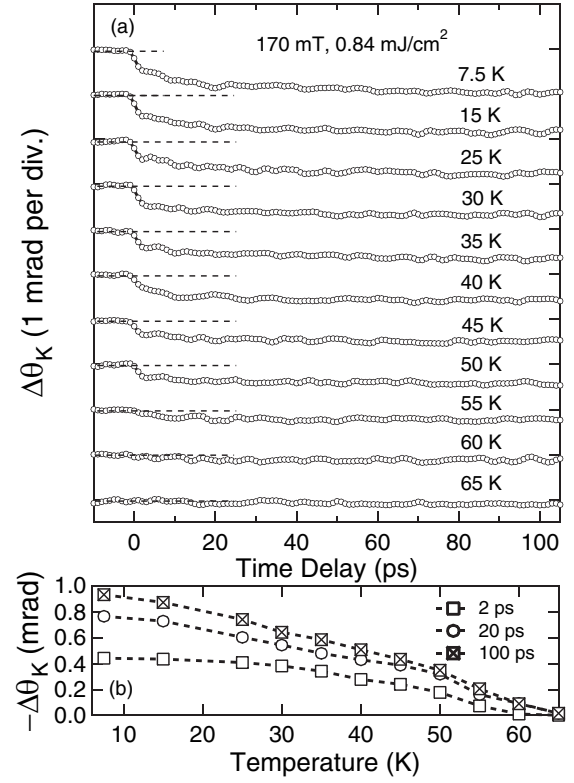

FIG. 4. Photoinduced MOKE angle change (a) versus time for different temperatures and (b) versus temperature for different time delays. The photoinduced change is absent above the Curie temperature (~60 K).

In the standard picture of laser-induced demagnetization, the heat deposited in the lattice is transferred into the spin system through spin-lattice interaction (for a microscopic model, see, e.g., [18]). The heating up of the spin system then results in demagnetization, occurring at the time scale of spin-lattice relaxation, τ_{sl}, which is at least of the order of 100 ps (≈ 80 ps in Gd [19] and over nanoseconds in paramagnetic CdMnTe [20]). Much faster demagnetization is expected when the process of direct carrier-spin interaction is taken into account. Such an interaction has been identified as the source of possible fast demagnetization in metals [2], and the importance of the spin-orbit scattering of carriers has been stressed [6].

Here, we can put these ideas on a stronger footing, exploiting the simplicity of carrier-spin coupling in diluted magnetic semiconductors (DMS). The p-d exchange interaction $H_{p-d} \sim \beta \mathbf{S} \mathbf{s}$ (see, e.g., [21]) couples the spins of delocalized holes with localized Mn moments; the latter are the main source of the macroscopic magnetization $\mathbf{M}$ (the carrier contribution to $\mathbf{M}$ is very small). The mean-field part of this interaction causes a spin splitting of bands proportional to the average value of Mn spin, which is exactly what is measured in magneto-optical experiments. Photoexcitation creates a nonequilibrium population of hot holes, with a blurred Fermi surface. This strongly increases

the number of states available for spin-flip scattering, coming from the off-diagonal part of H_{p-d}, allowing the flow of energy and angular momentum between carriers and localized spins. Demagnetization is caused by this flow of polarization from Mn to holes, which is sustained by efficient hole spin relaxation. The whole process can be envisioned as the reverse of the Overhauser effect: the excited carriers are becoming dynamically polarized at the expense of the localized spins, and the dissipation of magnetization occurs through spin relaxation in the carrier system.

Spin-flip scattering has been used to describe the cw heating of Mn by electrons in a paramagnetic DMS [22], and the idea of magnetization relaxation through spin flips with carriers with subsequent spin relaxation by spin-orbit interaction can be traced back as far as the 1950s [23] (recently rejuvenated for DMS [24,25]).

At lowest pump fluences the demagnetization is ~20% after 200 fs, and using the estimate of $p \simeq 4 \times 10^{20}$ cm^{-3} we see that every hole has to participate in ~1 spin flip $[0.2 \times \frac{5}{2} \times 10^{21}/(4 \times 10^{20})]$ to achieve such an effect. This leads to an upper bound for spin-relaxation time $\tau_s \lesssim$ 200 fs, which is reasonable: the momentum scattering time in disordered III-V DMS is of the order of 10 fs [26], and furthermore, in the excited population other scattering mechanisms (carrier-carrier and phonon scatter-

ing) play a significant role. Strong spin-orbit interaction in the valence band leads to close correlation between the momentum scattering and spin-relaxation times.

In order to substantiate the above physical picture, we used a single heavy-hole spin-split band to calculate the spin-flip scattering rates [22]:

$$W_{m,m'} = \frac{2\pi}{\hbar} \frac{\beta^2}{4} \sum_{\sigma,\sigma'} \int \frac{d^3k}{(2\pi)^3} \int \frac{d^3q}{(2\pi)^3} f_{\sigma'}(\mathbf{k})(1 - f_\sigma(\mathbf{q}))$$
$$\times \delta(\epsilon_\sigma(\mathbf{q}) - \epsilon_{\sigma'}(\mathbf{k}) \pm \delta)|\langle m\sigma|\hat{S}_\pm \hat{\sigma}_\mp|m'\sigma'\rangle|^2, \tag{1}$$

where $m = -\frac{5}{2}, \ldots, \frac{5}{2}$, δ is the mean-field splitting of the Mn spin levels, and σ is the hole spin. The occupation functions $f_\sigma(\mathbf{k})$ are thermal, with a high effective temperature T_h mimicking the highly nonequilibrium state of holes, and with possible different Fermi levels for two spin directions. The dynamics of Mn spins is described by a simple rate equation for diagonal elements of the localized spin density matrix $\dot{N}_m = \sum_{m'}(W_{mm'}N_{m'} - W_{m'm}N_m)$, and the dynamics of the average hole spin s is given by $\dot{s} = -\gamma\dot{M} - \frac{1}{\tau_s}(s - s_{eq}(M, T_h))$, where M is the average Mn spin, γ is the ratio of Mn to hole density, and s_{eq} is the instantaneous equilibrium value of the average hole spin.

The results of the calculations show a demagnetization of 10% within 200 fs for $T_h = 1000$ K, $\tau_s = 10$ fs, and $p = 4 \times 10^{20}$ cm^{-3}, which compares well with the experiment. For much less favorable values of hole spin-relaxation time $\tau_s = 100$ fs, we get $\Delta M/M \approx 5\%$. More theoretical work on strong photoexcitation of disordered III-V DMS and spin relaxation of hot holes is needed to verify the parameters used above. However, it is clear that even our simple model shows that ultrafast demagnetization is possible in these materials.

The observed long-time dynamics is connected with the "traditional" pathway of heat transfer (with the intervention of the lattice). The results are similar to those seen by Kojima *et al.* [27] in GaMnAs, which were modeled by the three-temperature model [2,28]. Our model concentrates on the subpicosecond time scale, where we emphasize the role of polarization transfer between the systems (with the possibility of "bottleneck" when τ_s is not small enough), not only the heat exchange.

In conclusion, we have made the first observation of two distinct demagnetization regimes in the dynamics of laser-excited InMnAs. We interpret the novel demagnetization dynamics as a result of ferromagnetic exchange couplings and spin-lattice interactions, manifesting themselves as a fast (<1 ps) and relatively slow (~100 ps) components, respectively. The fast component completely quenches ferromagnetism at high pump fluences. Systematic power and temperature dependence of this ultrafast demagnetiza-

tion provides convincing evidence supporting the proposed physical picture.

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