Ultrafast Photoinduced Softening in a III-V Ferromagnetic Semiconductor for Non-thermal Magneto-Optical Recording

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Through time-resolved two-color magneto-optical Kerr spectroscopy we have demonstrated that photogenerated transient carriers decrease the coercivity of ferromagnetic InMnAs at low temperatures. This transient “softening” persists only during the carrier lifetime (∼2 ps) and returns to its original value as soon as the carriers recombine to disappear. We discuss the origin of this unusual phenomenon in terms of carrier-enhanced ferromagnetic exchange interactions between Mn ions and propose an entirely nonthermal scheme for magnetization reversal.

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Mn-doped III-V semiconductors offer a variety of new scientific and technological opportunities through their carrier-induced ferromagnetism \([1, 2, 3, 4, 5]\). Their magnetic characteristics are sensitive functions of carrier density, and hence, carrier-density-modulation can lead to control of ferromagnetism. In fact, voltage-tuned \([6, 7]\) and photoinduced \([8, 9, 10]\) alteration of ferromagnetism have been demonstrated recently. In parallel to these pioneering studies on ferromagnetic semiconductors are efforts to manipulate ferromagnetic metals (or itinerant ferromagnets). A number of recent time-domain optical studies of itinerant ferromagnets have revealed novel phenomena, including ultrafast demagnetization \([11]\). However, there have been few time-dependent optical studies of (III,Mn)V ferromagnets.

Here, we report on a demonstration of ultrafast photoinduced softening (UPS) (i.e., a transient photoinduced decrease of coercivity) in ferromagnetic InMnAs/GaSb heterostructures. Pumping (III,Mn)V systems with ultrashort laser pulses has a multitude of effects that are not present in the case of ferromagnetic metals. Since laser pulses can create a transient distribution of delocalized carriers, one can anticipate significant modifications in the exchange interaction between localized Mn spins and delocalized carrier spins. This should be contrasted to the metal case where the main effect of the laser pulses is heating (either electronic or lattice heating). We observed that photogenerated transient carriers significantly decrease the coercivity \((H_c)\). We attribute this phenomenon to the carrier-enhanced Mn-Mn exchange interaction and propose a new, extremely fast scheme for recording information on a magneto-optical disk entirely non-thermally.

Figure 1 shows a schematic band diagram of the InMnAs/GaSb sample studied. Surface pinning of the Fermi energy and the type-II broken-gap alignment between InMnAs and GaSb produces large band bending. Also shown in Fig. 1 is the selective pumping scheme we used. At the pump wavelength (2 μm), the photon energy (0.62 eV) was smaller than the band gaps of GaSb (0.812 eV) and GaAs (1.519 eV) but larger than that of InMnAs (∼0.42 eV), so the pump created carriers only in the InMnAs layer (Fig. 1). Under our pumping conditions, the maximum density of photocreated carriers is estimated

![Diagram](attachment:image.png)
to be comparable to or larger than the background carrier density ($\sim 10^{19}$ cm$^{-3}$), and, hence, significant modifications in exchange interactions can be expected.

We performed two-color time-resolved magneto-optical Kerr effect (MOKE) spectroscopy experiments using femtosecond ($\sim 150$ fs) pulses of midinfrared (MIR) and near-infrared (NIR) radiation. Details of the setup were described previously [12]. The source of intense MIR pulses was an optical parametric amplifier (OPA) pumped by a regenerative amplifier (Model CPA-2010, Clark-MXR, Inc.). The OPA was tunable from 522 nm to 20 $\mu$m using different mixing crystals. We used a very small fraction ($\sim 10^{-5}$) of the CPA beam (775 nm) as a probe and the output beam from the OPA tuned to 2 $\mu$m as the pump. The two beams were made collinear by a non-polarizing beam splitter and then focused onto the sample mounted in a magnet with optical windows. We recorded the intensity difference of the $s$- and $p$-components of the reflected NIR beam as well as its total intensity as functions of time delay and magnetic field.

The sample studied was an InMnAs/GaSb single heterostructure with a Curie temperature of 55 K, consisting of a 25 nm thick In$_{0.01}$Mn$_{0.99}$As magnetic layer and an 820 nm thick GaSb buffer layer grown on a semi-insulating GaAs (100) substrate. Its room temperature hole density and mobility were $1.1 \times 10^{19}$ cm$^{-3}$ and 323 cm$^2$/Vs, respectively. The magnetization easy axis was perpendicular to the epilayer due to the strain-induced structural anisotropy caused by the lattice mismatch between InMnAs and GaSb. This allowed us to observe ferromagnetic hysteresis loops in the polar Kerr configuration.

Typical data showing the ultrafast photoinduced softening process are presented in Figs. 1(b) and 1(c). Here, two magnetic-field scans exhibiting ferromagnetic hysteresis loops at 20 K are shown, for $-4$ ps and 430 fs time delays, respectively. The pump beam was circularly polarized. The data at $-4$ ps delay [Fig. 1(b)] shows a hysteresis loop with a finite coercivity ($\sim 1$ mT). However, at 430 fs [Fig. 1(c)], the hysteresis loop is suppressed in the horizontal direction, i.e., the coercivity is almost zero. It is important to note that the data shows almost no change [or a slight increase (i.e., $10\%$, if any) in vertical size, within the sensitivity of our setup]. This leads us to conclude that simple lattice heating cannot be the reason for the above observations since raising the lattice temperature should result in loop shrinkage both horizontally and vertically.

Figure 2 plots detailed photoinduced magnetization dynamics. Figure 2(a) shows photoinduced MOKE signal versus time delay at a magnetic field of $-20$ mT with a circularly polarized pump. An ultrafast photoinduced response is clearly observed. Ferromagnetic loops are plotted in Figs. 2(b), 2(c), 2(d), and 2(e), which correspond to the fixed time delays indicated in Fig. 2(a). Again, near time zero [Fig. 2(c)], the loop is collapsed horizontally. It can be also seen that such softening lasts only for a very short time, $\sim 2$ ps. As soon as the photoinduced MOKE signal disappears [see Fig. 2(a)], the loop returns with the original $H_c$ recovered [see Figs. 2(d) and 2(e)].

First, let us discuss the origin of the ultrafast signal in Fig. 2(a). Note that none of the field scans in Figs. 1 and 2 are offset; i.e., vertical shifts of the loops are a real effect. However, we observed similar ultrafast photoinduced MOKE signals even at temperatures far above the Curie temperature. Therefore, we believe that the ultrafast vertical shifts shown in Fig. 2(a) are not due to any change related to ferromagnetism. We can also exclude nonlinear optical effects such as state filling, band filling, and band gap renormalization as the cause of the induced MOKE signal. The two-color nature of the current measurements allows us to decouple the photogenerated car-
riers from the energy levels probed, i.e., the quasi Fermi level of the optically excited carrier system is too low to affect the probe. Finally, we found that the sign of the ultrafast MOKE signal depends on the sense of circular polarization of the pump pulse, i.e., $\sigma^+$ or $\sigma^-$. These facts and considerations lead us to conclude that the coherent spin polarization of the photogenerated carriers is the origin of the ultrafast photoinduced vertical shift of the ferromagnetic hysteresis loop. Thus, such time scans provide a direct measure of the spin lifetime of photogenerated carriers. In addition, we determined the charge lifetime of the carriers to be $\sim 2$ ps by standard pump-probe techniques. The short charge lifetime is probably due to anti-site defects introduced during low temperature MBE growth [13], but more systematic studies are necessary to understand the microscopic mechanism of the ultrafast carrier recombination dynamics.

We believe that carrier-enhanced exchange coupling between Mn ions is at the core of the observed ultrafast photoinduced softening. The phenomenon is essentially the same as what has been observed in the CW work on the same systems [2, 14, 15] except for the very different time scales. One possible scenario is in terms of domain walls [6]. Namely, creation of a large population of transient holes breaks the original balance between the exchange energy and the anisotropy energy (the latter was found to be independent of carrier density [8]). The dominance of the former results in an increase in domain wall thickness and a decrease in domain wall energy. This reduces the magnetic field required to achieve magnetization reversal, i.e., coercivity is decreased. Another possible mechanism involves the magnetic rotation of single-domain particles known as magnetic polarons. In an assembly of such particles, the interaction field among the particles is opposite to the particle magnetization and thus helps to reverse the particles. This gives rise to a reduction of the coercive field needed for magnetization reversal compared to that of a single particle. A large enhancement of the exchange coupling leads to the enlargement of magnetic polarons and thus increases the single particle magnetization. This increases the interaction field and reduces the coercive force. In either view, as soon as the transient photogenerated carriers recombine to disappear, the original value of the coercive field is recovered. This is consistent with the notion that the extremely short-lived photogenerated carriers are the cause of ultrafast photoinduced softening.

Finally, we propose a new scheme for nonthermally recording information onto a magneto-optical disk based on the observed ultrafast softening (Fig. 3). If a storage medium made of a III-V ferromagnetic semiconductor is prepared in a magnetic field that is smaller than the original coercivity but larger than the expected photo-modified coercivity ($H_{c,\text{mod}}$), ultrafast creation of transient carriers can cause magnetization reversal. Our experiments above showed that the softening process happens and persists only for less than 2 ps, after which the coercivity recovers its original value. Within such a time scale, any heat transfer process between spin and lattice subsystems, which takes more than 1 ns, is out of the picture. This proposed scheme, being nonthermal, possibly exceeds the fundamental limit to the data writing rate set by the thermo-magnetic process used in conventional magneto-optical recording (which is 30 MB/s in the state-of-the-art technology) [16].

In summary, we have demonstrated ultrafast photoinduced softening, i.e., a transient coercivity decrease, in an InMnAs/GaSb ferromagnetic semiconductor heterostructure. We found surprisingly fast switching due to ultrashort carrier lifetimes in ferromagnetic semiconductor InMnAs, exceeding the fundamental time limitation set by the thermo-magnetic process. The extremely fast decay and recovery of the observed ferromagnetic softening process differentiates itself from any known ultrafast processes in ferromagnetic systems. We attribute the observed softening to the carrier-enhanced exchange interaction between Mn ions and believe that further systematic studies will provide detailed information on the microscopic mechanism of carrier-mediated ferromagnetism in these semiconductors.

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