Ultrafast Enhancement of Ferromagnetism via Photoexcited Holes in GaMnAs

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We report on the observation of *ultrafast photo-enhanced ferromagnetism* in GaMnAs. It is manifested as a transient magnetization increase on a 100-ps time scale, after an initial sub-ps demagnetization. The dynamic magnetization enhancement exhibits a maximum below the Curie temperature T_c and dominates the demagnetization component when approaching T_c. We attribute the observed ultrafast collective ordering to the *p-d* exchange interaction between photoexcited holes and Mn spins, leading to a correlation-induced peak around 20K and a transient increase in T_c.

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There has been long and intense interest in searching for possibilities for *ultrafast enhancement* of collective magnetic order via photoexcitation. Such photoexcitation would lead to fascinating opportunities both for establishing a transient cooperative phase from an uncorrelated ground state and for determining the relevant time scales for the build-up of order parameters. Previous time-resolved investigations in paramagnetic II-VI semiconductors provided evidence for collective photoexcitations that led to the formation of magnetic polarons [1]. Substantial recent progress has been made in observing ultrafast spin reorientations, e.g., in antiferromagnetic materials [2, 3]. However, most prior experiments in magnetically-ordered materials only show an *ultrafast decrease* of the magnetization amplitude due to laser induced electronic heating [4]. Recently, several experiments in strongly correlated manganites and transition metal alloys revealed transient photo-induced magnetization on ultrafast time scales, but only *demagnetization* could be seen at temperatures away from T_c. The role of ultrafast pumping is most likely a *thermal* perturbation of competing phases near critical points [5, 6].

The discovery of hole-mediated ferromagnetism in III-V ferromagnetic semiconductors (III-V FMSs) such as GaMnAs and InMnAs [7] offers unique opportunities and flexibility for *nonthermal* control of magnetism. Unlike other types of magnets, the ferromagnetic exchange between localized Mn moments is mediated by free hole spins through *p-d* interaction $H_{p-d} \sim J_{pd}\mathbf{S}\cdot\mathbf{s}$ ($J_{pd} \sim 1$ eV), making the magnetic properties a sensitive function of the hole density. For instance, the trend of experimental T_c in GaMnAs is shown to be proportional to $p^{1/3}J_{pd}^2$, for a wide range of hole densities p [8]. Recently, hole-density-tuning via external stimuli such as CW light excitation [9] and electrical gating [10] has demonstrated clear enhancement of magnetization and increase of T_c, as illustrated in Fig. 1(a). However, no time-resolved experiments in III-V FMSs have shown transient photo-induced magnetization, and the time scale for the enhancement of collective order is completely unknown. So far only ultrafast demagnetization and quenching dynamics were observed, due in part to the relatively high pump fluences used (~ 1 mJ/cm^2) [11, 12]. Moreover, recent theory has pointed to the critical role of the Mn-hole exchange correlation in *ultrafast, nonthermal* manipulation of magnetization in GaMnAs [13], but no experimental evidence for this has been reported. In addition to their fundamental scientific interest, III-V FMSs are among the most promising candidates for future "multifunctional" devices and for quantum information technology based on the spin degree of freedom [14]. It is thus of significance to explore ferromagnetic enhancement in III-V FMSs to technologically important, sub-ns time scales.

In this Letter, we report the observation of *ultrafast enhancement of ferromagnetism* in GaMnAs. Our data clearly show photo-induced magnetization on a 100-ps time scale after initial sub-ps demagnetization. The dynamic magnetization enhancement exhibits a maximum below T_c and dominates the demagnetization component when approaching T_c. Our analysis and theoretical simulations based on the H_{p-d} interaction between photoexcited holes and Mn spins explain the salient features of the experiment showing, in particular, a correlation-induced peak around 20K and a transient increase in T_c.

The sample studied in the present work was a GaMnAs/GaAs heterostructure with a Curie temperature of 77 K. The sample was grown by low-temperature molecular beam epitaxy (MBE) and consisted of a 73-nm $Ga_{0.925}Mn_{0.075}As$ layer deposited on a GaAs buffer layer and a semi-insulating GaAs (100) substrate. The background hole density was approximately $3\cdot10^{20}$ cm^{-3}. Our measurements involved ultraviolet (UV) pump/near-infrared (NIR) magneto-optical Kerr effect (MOKE) spectroscopy. The experimental setup consisted of a femtosecond oscillator with 120 fs pulse duration and a BBO crystal in the pump path for doubling the photon energy. The pump beam was at 3.1 eV and was linearly polarized, with peak fluences $\simeq 10\mu$J/cm^2. Fig. 1(b) illustrates that fs pump pulses create a large density of holes in the valence band of GaMnAs. A small frac-

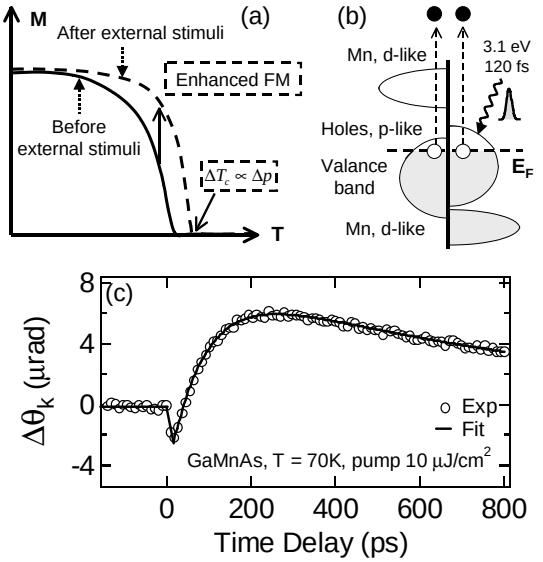

FIG. 1: (a) Illustration of hole-density-tuning effects via external stimuli in III-V FMSs seen in the static experiments [9, 10]. FM: ferromagnetism. $\triangle p$ is hole density change. (b) Schematic diagram of the spin-dependent density of states in GaMnAs. fs pump pulses create a transient population of holes in the valence band. (c) Time-resolved MOKE dynamics at 70K and under 1.0T field. Transient enhancement of magnetization, with $\sim$ 100 ps rise time, is clearly seen after initial fast demagnetization. Thick line is the fit described in the text.

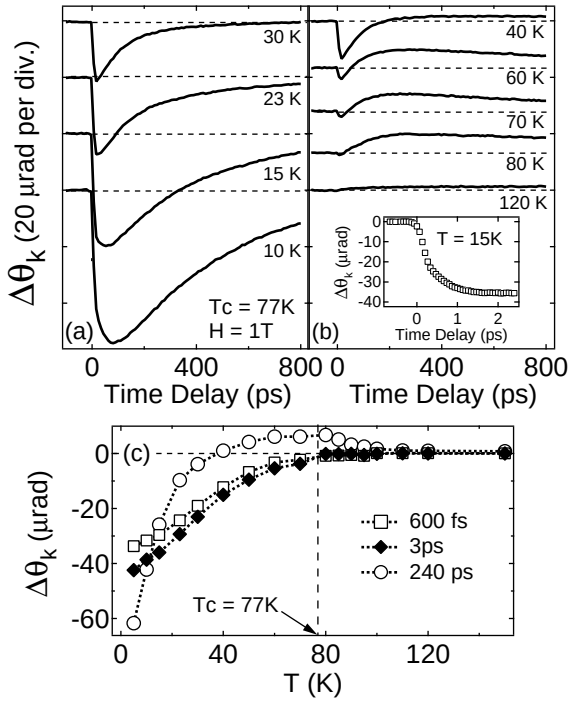

FIG. 2: (a)-(b) Temporal traces of photoinduced magnetization changes at different temperatures. All traces are intentionally offset for clarity. Inset: the first 2 ps demagnetization dynamics at 15 K. (c) Temperature dependence of magnetization changes at different time delays - 600 fs (squares), 3 ps (diamonds) and 240 ps (circles), respectively.

tion of the fundamental beam at 1.55 eV was used as a probe, detecting magnetization via the polar MOKE angle θ_K[15]. The low pump peak fluences and the high pump photon energy minimize spurious effects such as two-photon absorption and pump scattering. Additionally, the "magnetic origin" of the transient MOKE response is confirmed by separate measurements showing the overlap of the pump-induced rotation and ellipticity through the entire time scan range [16].

A typical temporal profile of transient MOKE changes $\Delta\theta_K$ at 70K is shown in Fig. 1(c), with a field of 1.0T perpendicular to the sample surface to align the magnetization. Two mutually competing dynamic magnetization processes are observed: an initial sub-ps demagnetization ($\Delta\theta_K < 0$), followed by a distinct magnetization rise on a 100 ps time scale ($\Delta\theta_K > 0$). The two processes clearly show different temperature dependences, as shown in Fig. 2(a)-(b). At elevated lattice temperature, the 200 fs demagnetization components [inset of Fig. 2(b)] quickly diminish and nearly disappear above T_c. This is also seen in the 600 fs and 3 ps traces in Fig. 2(c). More intriguingly, an enhancement of the transient magnetization begins to dominate the demagnetization component at high temperatures. For instance, the net magnetization changes become positive above 40K at long time delays, e.g., at 240 ps [Fig. 2(c)]. The photoinduced magnetization persists above T_c - as is clearly visible in the 80K trace [Fig. 2(b)] - and gradually vanishes at higher temperatures.

MOKE signals, measured with the 1.55 eV probe, arise from the macroscopic magnetization **M** - average localized Mn spins - through the coupling of the spin-split electronic states near the band edge. The background carrier spin contribution to **M** is negligible, and photoexcited transient carriers are not spin polarized since the pump contains no net angular momentum. Thus the positive MOKE signals, rising on a 100-ps time scale, clearly indicate an ultrafast alignment of Mn spins and an enhancement of ferromagnetic order. Our results reveal the ultrafast time scale of this process, not accessible in previous static measurements [9, 10].

We attribute the observed *ultrafast photo-enhanced ferromagnetism* to the transient hole-Mn interaction via the H_{p-d} exchange, as follows. At early pump-probe delays ($\triangle t \sim 0$ fs), the ultrashort laser pulses generate a nonequilibrium distribution of spin-unpolarized electron-hole pairs in GaMnAs under a finite external H field. During the first ps ($\triangle t < 1ps$), the photoexcited hot holes will experience efficient spin-flip scattering with the localized Mn moments, manifesting this as a sub-ps demagnetization component. This results from the off-diagonal elements of the exchange Hamiltonian H_{p-d} ($\sim J_{pd}\mathbf{S}_{\pm} \cdot \mathbf{s}_{\mp}$), which cause the spin polarization of the

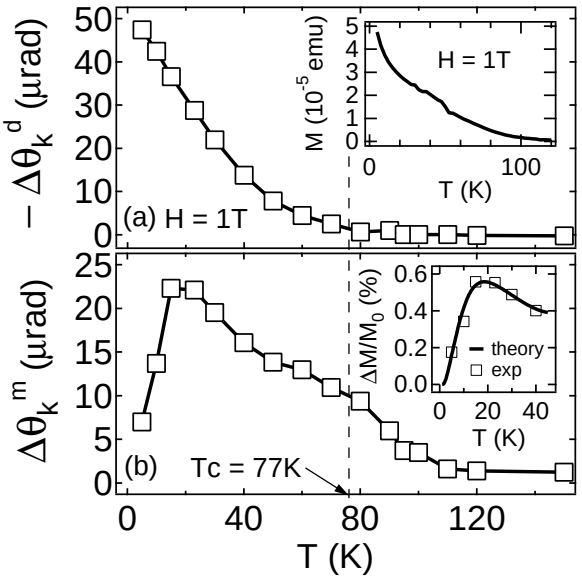

FIG. 3: Decomposed demagnetization $-\triangle\theta_k^d$ [(a)] and enhanced magnetization $\triangle\theta_k^m$ [(b)] components are plotted as a function of temperature. The static magnetization curve under 1.0T field is shown as inset in (a). The simulation $\triangle M/M_0$ (thick line, $\triangle T_c = 1.1K$ and the hole-Mn ratio of 0.06) and experimental values (circles, normalized by static θ_k at 5K) of the photo-enhancement peak $\sim$ 20K are shown as inset in (b).

Mn ions to transfer to the holes within several 100s of fs, similar to the fs demagnetization first reported in InMnAs [11]. Meanwhile, the hot hole distribution quickly cools via carrier-phonon scattering (optical phonon energy $\sim$36 meV), resulting in a rapid termination of demagnetization (within the first ps). At longer pump-probe delays of $\triangle t > 1ps$, the photoexcited, thermalized holes, settling down in the spin-split bands, can now participate in the process of hole-mediated ferromagnetic ordering. These *extra* holes enhance the Mn-Mn exchange correlation and polarize Mn spins via the mean-field (diagonal) elements of the exchange Hamiltonian H_{p-d} ($\sim J_{pd}\mathbf{S}_z \cdot \mathbf{s}_z$), thereby increasing the macroscopic magnetization.

In order to elucidate the salient features of the *photo-enhanced ferromagnetism*, we decompose the transient MOKE changes shown in Fig. 2 into demagnetization ($-\triangle\theta_k^d$) and enhanced magnetization ($\triangle\theta_k^m$) components, based on their different time scales. The temporal profile of $\triangle\theta_k$ is well described by $\triangle\theta_k^d \cdot exp(-t/\tau_d) + \triangle\theta_k^m \cdot (1 - exp(-t/\tau_m)) \cdot exp(-t/\tau_c)$. Here $-\triangle\theta_k^d$ and τ_d in the first term are the magnitude of demagnetization and the recovery time determined by the slow heat diffusion process, respectively. In the second term, $\triangle\theta_k^m$ and τ_m are the magnitude and build-up time of the enhanced magnetization component, respectively, while τ_c accounts for the final decay of the magnetization enhancement via hole diffusion and recombination. The time constant τ_c is on the order of a few ns, as seen in the decay of the positive MOKE signal. As an example, the thick line in Fig. 1(c) represents the fit of the MOKE dynamics at 70K.

Fig. 3 plots the temperature dependence of $-\triangle\theta_k^d$ and $\triangle\theta_k^m$. The $-\triangle\theta_k^d$ profile resembles the static magnetization curve of the sample, exhibiting strong deviation from the classical mean-field convex shape [inset, Fig. 3(a)]. This non-classical behavior of magnetization arises from the existence of two strongly interacting spin ensembles, Mn and holes, as discussed in [17]. More intriguingly, the magnetization enhancement $\triangle\theta_k^m$ in Fig. 3(b) shows a distinctly different temperature profile, with a peak of $\sim$ 0.5% of the saturation magnetization M_0 around 20K (static θ_k at 5K $\sim$ 4 mrad) and a prolonged tail extended to $\sim$ 120K. The extended profile beyond T_c is expected as the combined effect of an applied external field of 1.0T and of hole-enhanced magnetic susceptibility in the paramagnetic state. The most salient feature of the *photo-enhanced ferromagnetism* is the peak near 20K, which is a manifestation of the Mn-hole correlation H_{p-d}. This can be qualitatively understood as follows: the ferromagnetic molecular field acting on the Mn ions (holes) is determined by the average spin polarizations of the holes (Mn ions) via the Mn-hole exchange coupling J_{pd}. The effective field acting on the holes is much larger than that acting on the Mn ions, because of the large density of Mn ions compared to the holes. As a consequence, the hole polarization will remain saturated at a temperature T_h much higher than that needed for saturating the Mn magnetization ($T_{Mn}\sim$0). As the lattice temperature increases above T_{Mn} but less than T_h, the Mn spins with partial alignment begin to be efficiently polarized via photoexcited holes with near-unity magnetization. However, as the lattice temperature rises higher than T_h, because of reduced hole polarization the efficiency of this magnetization enhancement process quickly drops, thus resulting in a magnetization enhancement maximum at some temperature on the order of T_h.

Next we present a simple theoretical calculation to simulate the Mn-hole correlation-induced peak around 20K. As we discussed, the hole-enhanced ferromagnetic correlation results in an increase of T_c ($\triangle T_c > 0$). We calculate the non-classical magnetization curve [inset, Fig. 3(a)] based on a modified Weiss mean-field model to take into account the H_{p-d} correlation [17]:

$$M(T,\Delta T_c)/M_0 = B_S[-3\frac{T_c+\Delta T_c}{T}\gamma S^* \times B_s[-3\frac{T_c+\Delta T_c}{T} \gamma^{-1}S^*\frac{M(T,\Delta T_c)}{M_0} + \frac{g_h\mu_B H}{k_B T}] - \frac{g_i\mu_B H}{k_B T}]$$

where $B_{S,s}(x)$ is the Brillouin function, γ is the square root of the hole and Mn density ratio $\sqrt{p/n_i}$, $g_i(g_h)$ is Mn(hole) g factor and S^* is given by $\sqrt{sS/(s+1)(S+1)}$. The Mn and hole spins, S and s, are 5/2 and 1/2, re-

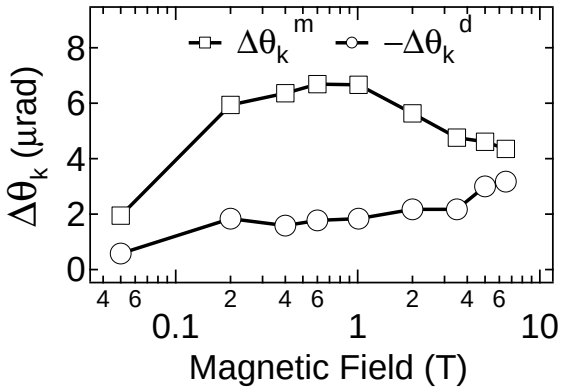

FIG. 4: Magnetic field dependence of photo-induced magnetization enhancement $\triangle\theta_k^m$ and demagnetization $\triangle\theta_k^d$ at 78K.

spectively. At each given $\triangle T_c$, the magnetization enhancement $\triangle M/M_0$ is solved self-consistently as a function of lattice temperature. The calculated temperature dependent $\triangle M/M_0$ and experimental $\triangle\theta_k^m$ (normalized by static θ_k at 5K) up to 40K are shown in the inset in Fig. 3 (b) for $\triangle T_c = 1.1K$. The results of the calculation compare well with the experimental photo-enhancement peak. In addition, we can also estimate $\triangle T_c$ using an analytical expression derived from the Zener Model, $\triangle T_c = \frac{1}{3}T_c \times \triangle p/p$ [18]. Knowing the ratio of photoexcited and background hole densities $\sim 2\%$, we estimate $\triangle T_c \sim 0.5$K, in reasonable agreement with the value used in the simulation.

The photoexcited transient electrons have little effect on $\Delta\theta_K$, because of the short free electron lifetime (< 1 ps) [19]. In addition, the standard thermal demagnetization via heat transfer from phonons, similar to the slow demagnetization component reported in [11, 12], is also seen at temperatures below 15K [Fig. 2(a)], which is not important to the physical picture discussed in this paper.

So far our discussion has been based on a situation involving a fixed magnetic field of 1.0T, which is important at low temperatures in order to eliminate other distractions (such as magnetization reorientation effects), without losing universality. It is worthwhile, however, to consider the field dependence of the magnetization enhancement at temperatures just above T_c and possible *transient* signatures of photo-induced critical phenomena, e.g., para- to ferromagnetic phase transition via the pump-induced increase of T_c. Fig. 4 presents the field dependence of the magnetization enhancement $\triangle\theta_k^m$ and demagnetization $-\triangle\theta_k^d$ at 78 K. Here the most interesting aspects lie in the fact that $\triangle\theta_k^m$ is relatively constant across a wide field range (0.2T - 1T) and only drops off close to zero T. It is crucial to note that, unlike static equilibrium measurements, *transient* pump-induced magnetization above T_c always reduces to zero at small external field, even though $\triangle T_c > 0$. A below-threshold H field is not able to activate subsequent growth of magnetic domains, even though they are nucleated via possible photo-induced long-range ferromagnetic correlation. Therefore, although the 1K increase of $\triangle T_c$ and the substantial transient magnetization enhancement at a small field of 0.05T suggest a transient photo-induced para- to ferromagnetic phase transition, more studies are needed to further elucidate the details of these transient features. Finally, the decrease (increase) of $\triangle\theta_k^m(-\triangle\theta_k^d)$ observed from 2T to 7T is expected from the larger static magnetization achieved at higher fields, resulting in the smaller photo-induced changes, as indeed shown by the data.

In summary, we have observed *ultrafast photo-enhanced ferromagnetism* in GaMnAs. Our data clearly show that the dynamic magnetization build-up occurs on a 100-ps time scale and exhibits a maximum below T_c. Our analysis and theoretical simulations, based on H_{p-d} interaction between photoexcited holes and Mn spins, explain the salient features of the experimental observations, demonstrating in particular a correlation-induced peak below T_c and a transient increase of T_c. Our measurements thus reveal a new transient collective magnetic phenomenon, and identify the critical role of *non-thermal* Mn-hole exchange correlation in this photo-induced cooperative behavior. The new functionalities in sub-ns time scales reported here may open future opportunities for high-speed spin-photon-charge integrated devices.

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