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## Interband dephasing and photon echo response in GaMnAs

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Coherent carrier dynamics are studied in GaMnAs using time-integrated and time-resolved four-wave mixing techniques. Dephasing is observed to be dominated by spin-flip scattering between the optically injected holes and Mn ions, revealing the rapid time scale of this scattering process in the III-Mn-V diluted magnetic semiconductors. The optical response is shown to exhibit the characteristic signatures of a simple photon echo, despite the complexity of band tail contributions and strong exchange coupling in this system. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4742916>]

Diluted magnetic semiconductors (DMS) are promising for applications in opto-electronics due to their large magneto-optical response<sup>1–4</sup> and the carrier-mediated nature of ferromagnetic coupling.<sup>5–10</sup> The large Faraday effect is attractive for optical isolators that may be directly integrated with III–V semiconductor lasers.<sup>11–16</sup> Recent observations of optical control of ferromagnetic order on femtosecond time scales, including ultrafast demagnetization,<sup>17,18</sup> magnetization precession,<sup>19–25</sup> and optical control of the coercive field,<sup>26–28</sup> may lead to optically addressable memory elements for magneto-sensitive semiconductor devices.<sup>29–34</sup> For such applications, the linear and nonlinear optical response of these materials, including the dynamic evolution following femtosecond excitation, must be well characterized.

Considerable insight has emerged in recent years regarding the ultrafast magnetization response,<sup>17,18,20–25</sup> the electron spin relaxation kinetics,<sup>35</sup> and carrier trapping and recombination processes in DMS materials.<sup>18,36–39</sup> In contrast, very little is known about the time scale for decay of optically induced coherence in the carrier system,<sup>40</sup> despite the fact that the interband dephasing time ( $T_2$ ) is a key parameter for theories of coherent ultrafast manipulation of the magnetization in DMS materials.<sup>42,43</sup> In a two-pulse four-wave mixing experiment, pulses  $\vec{E}_1(t - t_1)$  and  $\vec{E}_2(t - t_2)$  propagating in slightly different directions (with wavevectors  $\vec{k}_1$  and  $\vec{k}_2$ ) excite a macroscopic polarization on the electronic transitions resonant with the laser spectrum. The four-wave mixing signal corresponds to emission along  $2\vec{k}_2 - \vec{k}_1$  generated by a component of this polarization, a process commonly referred to as self-diffraction. The resulting signal contains crucial information about the interactions that lead to coherence decay in the system of excited electron-hole pairs.<sup>46–49,51,52,54,55</sup> Measurements of  $T_2$  in CdMnTe quantum wells using four-wave mixing techniques have provided new insight into the exchange coupling between Mn dopants and carriers,<sup>41</sup> however, no measurements of  $T_2$  have yet been reported in DMS materials based on III–V semiconductors. Here, we present measurements of interband dephasing kinetics in GaMnAs. Our results indicate that spin-flip scattering between the optically excited holes and Mn ions provides the dominant dephasing mechanism, with an upper

bound on  $T_2$  of 40 fs for  $x \geq 0.014\%$ . Our experiments have thereby provided a direct measurement of the time scale of this scattering process in GaMnAs. Through detection of the time envelope of the four-wave mixing emission using femtosecond gating techniques, we also show that the optical response at the band edge of GaMnAs consists of a simple photon echo, despite the complexity of exchange coupling and defect-induced localization in this system.

The samples studied in this work consist of 800 nm of Ga<sub>1-x</sub>Mn<sub>x</sub>As grown on a semi-insulating GaAs substrate by molecular beam epitaxy. A stop etch layer (175 nm of Al<sub>0.27</sub>Ga<sub>0.73</sub>As) was deposited prior to the GaMnAs layer to permit removal of the substrate for experiments in the transmission geometry.<sup>50</sup> The substrate temperature during growth of the GaMnAs layer was 250 °C. The Mn content was determined using secondary ion mass spectrometry,<sup>44</sup> and varies from 0 (low-temperature-grown GaAs, LT-GaAs) to 0.1%. A reference sample of GaAs was also grown at a substrate temperature of 600 °C (high-temperature-grown GaAs, HT-GaAs) for comparison with the low-temperature grown samples. The optical source used in this work is a mode-locked Ti:Sapphire oscillator, producing pulses with a center photon energy of 1.49 eV. A prism compressor was used for dispersion compensation, resulting in a pulse duration at the sample position of 22 fs, measured using zero background autocorrelation techniques at an equivalent focus. The total excited electron hole pair density was estimated for each sample using the fraction of light transmitted. The optical density was measured to be  $\leq 0.2$  for all samples, indicating that propagation effects may be neglected. Our experiments were performed in the small signal limit ( $\chi^{(3)}$  regime), as verified from the cubic power dependence of the detected signal. For all experiments, the samples were held in a liquid helium flow cryostat at 10 K.

Results of time-integrated four-wave mixing (TI-FWM) experiments are shown as a function of the time delay between the two excitation pulses ( $t_2 - t_1$ ) in Fig. 1(a) for GaMnAs with  $x = 0.1\%$ . The corresponding results for HT-GaAs and the pulse autocorrelation are also shown. For these measurements, the four-wave mixing beam was passed through a monochromator (resolution 2 meV) prior to detection to

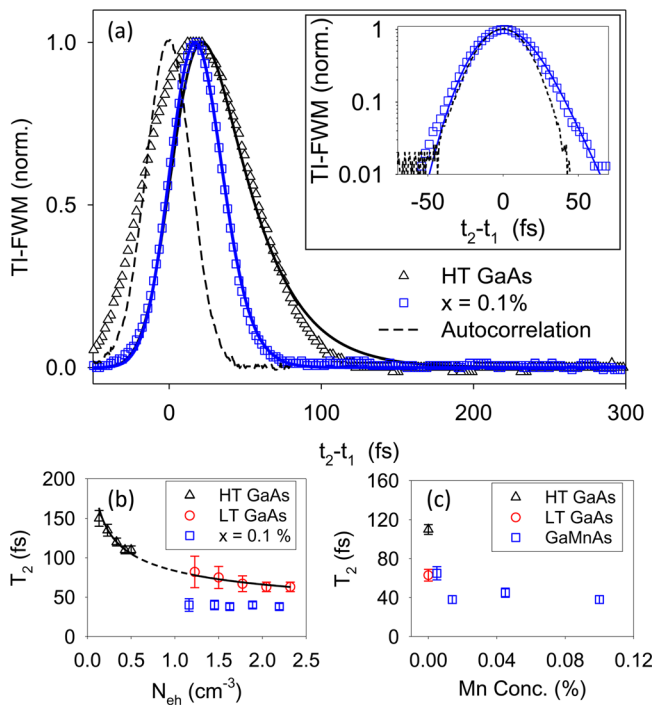


FIG. 1. (a) TI-FWM results at 10K for HT-GaAs (triangles) and  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  for  $x=0.1\%$  (squares), together with the pulse autocorrelation (dashed curve). These data were taken at a photon energy of 1.53 eV. The solid curves are fits to the formula in Ref. 45. Inset: results for  $x=0.1\%$  shown on a logarithmic scale after translating to zero delay for comparison with the pulse autocorrelation. (b)/(c): Interband dephasing times extracted from fits to the TI-FWM results as a function of (b) optically injected carrier density ( $N_{eh}$ ); and (c) Mn concentration. The values of  $N_{eh}$  are much larger for LT-GaAs and GaMnAs than HT-GaAs for similar pump beam powers due to absorption into band tail states (see Ref. 40). The solid curve in (b) indicates a fit of the data for HT-GaAs and LT-GaAs to  $\frac{1}{T_2} = aN_{eh}^{\frac{1}{2}}$ , with  $a=0.012$  cm/fs.

discriminate between the response of the exciton and continuum electron-hole pair states.<sup>40</sup> The data shown correspond to a photon energy of 1.53 eV ( $\sim 10$  meV above the band gap, in the interband continuum). The position of zero delay was determined using the mirror image of the signals along  $2\vec{k}_2 - \vec{k}_1$  and  $2\vec{k}_1 - \vec{k}_2$ , with an accuracy of  $\pm 10$  fs. For both HT-GaAs and GaMnAs, the TI-FWM signal reaches a maximum at positive time delay, however, the decay is much more rapid in the Mn-containing samples. The inset shows the results for  $x=0.1\%$  on a logarithmic scale, where the peak of the TI-FWM signal was translated to zero delay for comparison with the pulse autocorrelation. The decay of the measured TI-FWM response was found to be comparable to the time resolution of our experiments for  $x \geq 0.014\%$ .

To gain further insight into the nature of the optical response in GaMnAs, the time-envelope of the four-wave mixing signal was measured by upconverting the spectrally integrated signal pulse with a third (gate) pulse. The results of these time-resolved four-wave mixing (TR-FWM) experiments are shown in Fig. 2(a) for HT-GaAs and in Fig. 2(b) for GaMnAs ( $x=0.005\%$ ). The arrival times of the two excitation pulses ( $t_1$  and  $t_2$ ) were determined to an accuracy of  $\pm 5$  fs by upconverting the scattered laser light from the respective beams along the detection direction ( $2\vec{k}_2 - \vec{k}_1$ ) with the gate pulse. For HT-GaAs, the TR-FWM signal at zero delay exhibits a slow rise on a time scale of  $\sim 100$  fs,

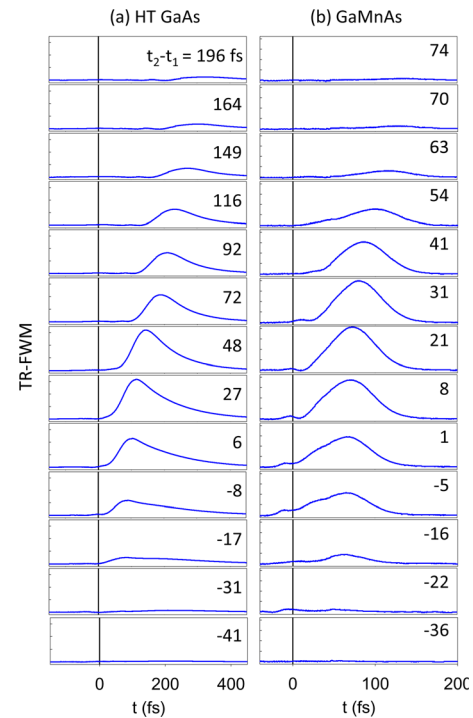


FIG. 2. TR-FWM results at 10K for (a) HT-GaAs and (b)  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  for  $x=0.005\%$  for various value of the time delay  $t_2 - t_1$ . The arrival time of pulse 1 ( $t_1$ ) is chosen as position of zero time  $t$ .

followed by an exponential decay with a time constant of 200 fs. Such a response, which is only present during overlap of the two excitation pulses, is consistent with an excitonic contribution mediated by exciton-carrier scattering, as seen in earlier experiments in HT-GaAs by several groups.<sup>51,52</sup> This excitonic contribution is removed by spectral filtering in the TI-FWM experiments described above. Beyond the region of pulse overlap ( $t_2 - t_1 \geq 35$  fs), the TR-FWM signal in HT-GaAs consists of a symmetric peak that shifts to larger times with increasing pulse delay. For the GaMnAs response in Fig. 2(b), the signal peak is symmetric for all time delays, and shifts towards larger times with increasing pulse delay, similar to the signal characteristics in HT-GaAs beyond the region of pulse overlap.

The general features of the four-wave mixing signals in HT-GaAs and GaMnAs are consistent with a photon echo response associated with an inhomogeneously broadened two-level system. From an analytical treatment in the short pulse limit,<sup>45</sup> such a photon echo response is manifest in TI-FWM experiments by a shift of the peak of the signal to positive time delay, in agreement with the results in Fig. 1(a). Physically, the position of the peak is determined by the degree of inhomogeneous broadening and the value of  $T_2$ . The two level model in Ref. 45 provides an excellent fit to the measured results after convolution with the Gaussian laser pulse profile, as shown by the solid curves in Fig. 1(a). The characteristic signature of a photon echo is the observation in TR-FWM of a peak close to  $t = 2(t_2 - t_1)$  after the arrival of pulse one (i.e., relative to  $t_1 = 0$ ) for delays that are long relative to the optical pulse duration.<sup>45</sup> Such a trend is observed in both HT-GaAs and GaMnAs, as shown in Fig. 2. Evidence for a photon echo response was observed previously in TR-FWM experiments in HT-GaAs.<sup>48,49</sup> For GaMnAs, the

optical response in the vicinity of the band gap is complicated by the presence of As antisites (introduced during low-temperature growth) and strong hole-Mn exchange coupling. The former leads to band tail transitions below the optical band gap, for which the degree of localization varies with carrier energy.<sup>53</sup> The latter is expected to increase the degree of inhomogeneous broadening due to coupling of holes to Mn ions with a random spin orientation,<sup>41</sup> and has been shown to enhance the valence band density of states.<sup>40</sup> Despite these complications, a linear shift in the peak position is observed beyond the region of pulse overlap (Fig. 3(b)) and the two level model provides excellent agreement with the shape of the TR-FWM trace at a fixed delay (solid curve in Fig. 3(a)). (It should be noted that the only free parameters in the model are the inhomogeneous bandwidth and the  $T_2$  time, which were both extracted from the TI-FWM results, as discussed in more detail below.) The observation of a simple photon echo response in GaMnAs indicates that the dephasing rate is independent of carrier energy despite the prevalence of band tail states and strong hole-Mn exchange coupling.

Fits to the TI-FWM response using the two-level model in Ref. 45 were used to extract  $T_2$  for all samples. In each case, the inhomogeneous bandwidth was determined from the width of the TI-FWM spectrum at zero delay. The resulting values of  $T_2$  are shown in Figs. 1(b) and 1(c). A strong dependence of  $T_2$  on the optically injected electron-hole pair density ( $N_{eh}$ ) was observed in both HT-GaAs and LT-GaAs. The density dependence is well described for both samples by  $\frac{1}{T_2} = aN_{eh}^3$ , with  $a = 0.012$  cm/fs, shown as the solid curve in Fig. 1(b). This density dependence is in agreement with

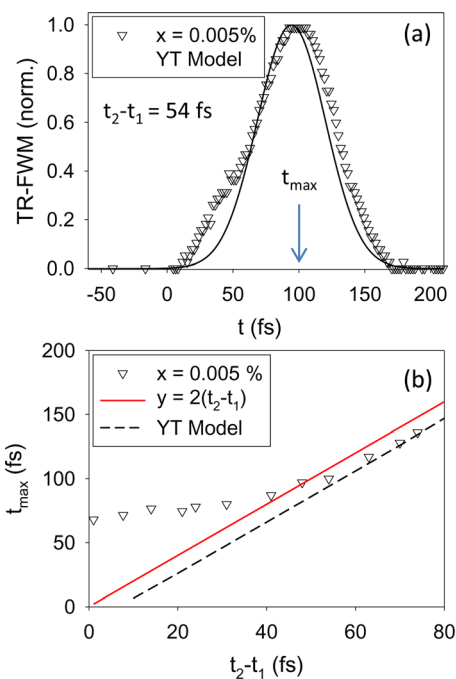


FIG. 3. (a) TR-FWM results at 10 K for  $x = 0.005\%$  at a pulse delay of 54 fs (triangles); solid curve: fit using the analytic result in Ref. 45 convoluted with the Gaussian laser pulse profile. The arrival time of pulse 1 ( $t_1$ ) is chosen as position of zero time  $t$ . (b) Peak position of the TR-FWM response relative to  $t_1 = 0$  for  $x = 0.005\%$  (triangles), together with the analytic result in Ref. 45 for the short-pulse limit (dashed curve). A similar trend was found for HT-GaAs (data not shown). The photon echo occurs slightly earlier than  $2(t_2 - t_1)$  (solid curve) due to the finite inhomogeneous broadening and dephasing time (see Ref. 45).

earlier four-wave mixing studies in GaAs,<sup>46–49</sup> in which the  $\frac{1}{3}$  exponent was attributed to carrier-carrier scattering in the presence of screening. (The importance of screening despite the short time scale of these experiments was established through comparison with quantum kinetic simulations by Hügel *et al.*<sup>49</sup>)

The absence of a density-independent dephasing contribution in LT-GaAs indicates that As-related defects introduced during low-temperature growth provide a negligible contribution to dephasing relative to carrier-carrier scattering for the conditions of our experiments. In contrast,  $T_2$  was observed to be independent of carrier density in  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  for all  $x$ , and was limited by the temporal resolution of our experiments for  $x \geq 0.014\%$  (Figs. 1(b) and 1(c)). Since carrier-carrier scattering and coupling to As-related defects are expected to contribute similarly in LT-GaAs and GaMnAs, the faster dephasing process in GaMnAs is attributed to the incorporation of Mn.

The sp-d exchange interaction leads to spin-flip scattering between the localized Mn spins and hole spins, a process that plays a crucial role in optically induced demagnetization in DMS systems.<sup>17</sup> This spin-flip scattering process also introduces an additional source of dephasing, accounting for the shorter  $T_2$  we observe in GaMnAs relative to LT-GaAs and HT-GaAs. Our experiments therefore reveal the rapid time scale of this process, indicating an upper bound of 40 fs in  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  for  $x \geq 0.014\%$ , providing new insight into exchange coupling in this system. The interband dephasing rate has been shown theoretically to have a critical influence on the optically induced magnetization trajectory in coherent control experiments in DMS materials.<sup>42</sup> Due to the potential for application of this ultrafast control process in optically addressable memory elements, knowledge of  $T_2$  is of crucial importance. The experiments we report here provide insight into the response of the carrier system in the femtosecond regime.

In summary, the interband dephasing kinetics in GaMnAs were investigated using femtosecond four-wave mixing techniques. A photon echo response is observed, indicating a uniform dephasing rate for the interband transitions despite the prevalence of band tail states in this system. Our experiments reveal the presence of a rapid dephasing process in GaMnAs, which we attribute to spin-flip scattering between the optically injected holes and Mn ions, providing a direct measurement of the time scale of this process. Our experiments provide insight into exchange coupling and ultrafast carrier dynamics in diluted magnetic semiconductors.

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