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We report ultrafast transient-grating measurements, above and below the Curie temperature, of the dilute ferromagnetic semiconductor (Ga,Mn)As containing 6% Mn. At 80 K (15 K), we observe that photoexcited electrons in the conduction band have a lifetime of 8 ps (5 ps) and diffuse at about 70 cm$^2$/s (60 cm$^2$/s). Such rapid diffusion requires either an electronic mobility exceeding 7700 cm$^2$/V s or a conduction-band effective mass less than half the GaAs value. Our data suggest that neither the scattering rate nor the effective mass of the (Ga,Mn)As conduction band differs significantly from that of GaAs. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4804578]

The dilute magnetic semiconductor (Ga,Mn)As is considered an archetypal spintronic material. Despite years of intensive study, the band structure of its holes continues to excite debate.1 The conduction band, on the other hand, is considered an archetypal spintronic material. Despite years of

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(001) semi-insulating GaAs. The structure consists of a 250 nm stop-etch layer of Ga$_{0.35}$Mn$_{0.65}$As, a 25 nm buffer of GaAs, and the 800 nm sample of Ga$_{0.95}$Mn$_{0.05}$As, grown with a Mn cell temperature of 760 °C and a substrate temperature of 215 °C. The film’s resistivity indicates a Curie temperature $T_C \approx 56$ K. From these data, we estimate the hole density as $p_0 \approx 1 \times 10^{20}$ cm$^{-3}$. After growth, the film was affixed to a sapphire window and the GaAs substrate removed by chemical etching.$^{10}$ Since

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Al$_{0.35}$Ga$_{0.65}$As is transparent at our measurement wavelength of 810 nm, our signal arises entirely in the (Ga,Mn)As film. The thickness of the (Ga,Mn)As film was chosen to ensure adequate signal.$^{11}$ We expect a high concentration of interstitial Mn defects, as films this thick cannot be effectively annealed.

To measure electron diffusion, we use an ultrafast transient-grating method.$^{12}$ A pair of “pump” laser pulses, split by a diffractive optic,$^{13,14}$ is simultaneously incident on the sample. The two pulses are non-collinear and interfere; their absorption excites photocarriers in a sinusoidal pattern with wavelength $\lambda$ and wavevector $q = 2\pi/\lambda$. By locally modifying the index of refraction, the carriers create a “grating” off of which time-delayed probe pulses diffract. As carriers recombine and diffuse, the grating amplitude decays at a rate

$$\frac{1}{\tau(q)} = D_\text{aq}^2 + \frac{1}{\tau_0}. \quad (1)$$

$D_\text{aq}$ is the ambipolar diffusion coefficient and $\tau_0$ is the lifetime of spatially uniform excitation. Measurement at several $q$ determines $D_\text{aq}$. Coulomb attraction constrains the electrons and excess holes to diffuse together, making diffusion ambipolar.$^{15}$ The motion is controlled primarily by the species with the lower conductivity.

We measure the diffracted probe amplitude in a reflection geometry, improve measurement efficiency by heterodyne detection,$^{16}$ and suppress noise by 95-Hz modulation of the grating phase and lock-in detection.$^{17}$ The laser pulses have wavelength near 810 nm, duration 120 fs, and repetition rate 80 MHz. The pump pulses have fluence at the sample of 4.2 $\mu$J/cm$^2$, and the probe pulses 0.35 $\mu$J/cm$^2$. At 810 nm GaAs has an absorption length$^{18}$ of order 1 $\mu$m and reflectivity of 0.3, so each pair of pump pulses photoexcites electrons and holes at a mean density of $n_\text{ex} \approx 1.2 \times 10^{13}$ cm$^{-3}$. The incident photons have energy about 30 meV greater than the bandgap of GaAs at 80 K. (Ga,Mn)As is known to suffer bandgap narrowing of order 120 meV.$^{19}$ So we estimate that our photoexcited electrons have excess energy of 150 meV. Screening of the electron-LO phonon interaction$^{20}$ could slow the electrons’ cooling to the lattice temperature, but
they will rapidly thermalize with the much more numerous holes through carrier-carrier scattering. The holes’ specific heat is about 100 times greater than the electrons’, resulting in a total heating of just 20 K. Thus, the rapid diffusion that we observe does not reflect the transport properties of hot electrons, but of equilibrium ones.21,22

Figure 1(a) shows the diffracted signal $S(t)$ as a function of time for gratings of several $q$. In each case, the signal is well described by the form

$$S(t) = Ae^{-t/\tau_{\text{fast}}} + Be^{-t/\tau(q)} + C,$$

with $\tau_{\text{fast}} \approx 0.34$ ps, $2.5$ ps $< \tau(q) < 8$ ps, $B/A = 0.11 \pm 0.06$, and $C/A = -0.012 \pm 0.007$. (The errors are standard deviations.) Of these three components of our signal, we will see below that the $\tau(q)$ term reveals the dynamics of free electrons diffusing at about 70 cm$^2$/s.

Previous ultrafast experiments on (Ga,Mn)As have observed signals with a very slow component corresponding to residual heat from the excitation pulse.26,27 Such experiments have also observed fast processes on the scale of $\tau_{\text{fast}}$ which were variously attributed to intraband relaxation or trapping of electrons at As antisite defects. Rapid trapping is consistent with our observation of electrons’ diffusion at later times: The number of electrons photoexcited likely exceeds the number of trapping sites, leaving some electrons untrapped and free to diffuse. Our observations contradict reports26 that all carriers are trapped and recombined within a picosecond.

Since $D_a$ is determined from $\tau(q)$, it is critical to find the latter values reliably, despite the constant and $\tau_{\text{fast}}$ terms, which introduce free parameters when fitting the data to Eq. (2). By measuring $S(t)$ at times much greater than $\tau(q)$, we determine $C$ unambiguously. Since $\tau_{\text{fast}}$ appears to have similar values at all $q$, we reduce parameters further by fitting all the data with the single value $\tau_{\text{fast}} = 0.34$ ps. The resulting values of $1/\tau(q)$ appear as the points in Figs. 2(a) and 2(b). This procedure gives reliable fits to the data’s $\tau(q)$ component, as evidenced by Figs. 1(b) and 1(c), in which we plot the measured $S(t)$ with the constant and $\tau_{\text{fast}}$ terms subtracted. The signal decays exponentially, and the decay is visibly faster at high $q$, indicative of diffusive motion.

Finally, we repeat each fit for a series of fixed values of $\tau(q)$ and judge “by eye” for what range of $\tau(q)$ the fits appear to plausibly reproduce the data. The outer limits of this range determine the error bars shown in Fig. 2(a).

The line in Fig. 2(a) is a fit to the form of Eq. (1), giving $D_a = 67$ cm$^2$/s and $\tau_0 = 8$ ps. (Fits to the lower or upper ends of the error bars give values of $D_a$ ranging from 60 to 80 cm$^2$/s, and $\tau_0$ from 14 to 6 ps.) For the 15-K data (Fig. 2(b)), $D_a$ is between 50 and 70 cm$^2$/s, and $\tau_0$ is between 3.4 and 5.2 ps. This figure makes it evident that our signal’s $\tau(q)$-component arises from ambipolar diffusion of free electrons, for the diffusivity is much faster than expected for heat or trapped carriers, while being comparable to prior measurements in $p$-type GaAs and InP.31

We note that we see much the same result on a sample with nominal Mn doping of 5% and 800-nm thickness, which gives $D_a$ of 53 cm$^2$/s at 15 K and 104 cm$^2$/s at 80 K (Fig. 2(c)). The temperature dependence may arise from the electrons’ mobility, or from spin-splitting of the conduction band below $T_C$. Our etching procedure succeeds rarely; most often, the sample disappears completely or the etched sample scatters too much light to allow measurements. For this reason the 5% sample is not etched, and the measured signal may include a small component from the GaAs substrate. Thus these measurements should be seen merely as corroboration of our result from the etched, 6% sample, which shows similar $D_a$ and similar variation with temperature.

Though the $\tau_0 = 8$ ps that we measure at 80 K may seem too short to be the free-electron lifetime, it is consistent with previous observations in low-temperature-grown GaAs and in (Ga,Mn)As, where a transient lasting 6 to 14 ps was attributed to non-radiative recombination of trapped carriers.35,36 Our signal arises from free electrons, not trapped, but is sensitive to recombination because, when all traps are filled, free electrons are trapped only as quickly as recombination empties traps. In what follows, $n_f$, the number of untrapped electrons, will be important. Our “best guess” is that $n_f/n_{ex} \approx B/A \approx 0.1$, i.e., that the relative amplitude of the $\tau(q)$ and $\tau_{\text{fast}}$ terms represents the proportion of photoexcited electrons remaining untrapped after 0.34 ps. Note, however, that our conclusions will hold true for the entire possible range $0 < n_f < n_{ex}$.

FIG. 1. Diffracted amplitude $S(t)$ for $q = 12000$ cm$^{-1}$ (circles), 37700 cm$^{-1}$ (squares), 45 300 cm$^{-1}$ (diamonds), and 52 800 cm$^{-1}$ (triangles). Scaled and offset vertically for clarity. Solid lines are fits to the form of Eq. (2). (a) Entire signal; semilog $t$. (b,c) Signal with constant and $\tau_{\text{fast}}$ terms subtracted, leaving just the $\tau(q)$ component; semilog $S$. 

FIG. 2. (a,b) Decay rate of the signal’s $\tau(q)$ component vs. $q^2$. Points and error bars are obtained by procedures described in the text. The lines are least-squares fits of the points to Eq. (1), revealing diffusion of photoexcited carriers. (c) Similar plots for a 5%-Mn, non-etched sample at 15 K (squares) and 80 K (circles). Errors are smaller than for the etched sample and can be estimated by the scatter of the points.
We now show that it is possible to use our measurements of $D_a$ and resistivity $\rho$ (of the holes) to infer conduction-band properties, despite our uncertain knowledge of three key parameters. The first is $\mu_e$, the electrons’ mobility; second is $n_i$. Finally, the electrons’ charge-susceptibility is $\chi_e = \partial n_e / \partial \mu_{chem}$, with $\mu_{chem}$ the chemical potential. This quantity approaches $n_i/k_B T$ at high temperature, and at low temperature approaches $N_e$, the electrons’ density of states at their quasi-Fermi energy; it thus provides insight into the electrons’ effective mass $m^*$. (With $J_b$ defined similarly for holes, the reduced susceptibility is $1/\chi_e \equiv 1/\chi_b + 1/\chi_h$. In this experiment, $\chi_b > \chi_e$ and $\chi_e \approx \chi_h$; we nonetheless include the hole susceptibility in our numerical calculations.)

By the Einstein relation, the electrons’ diffusivity is

$$D_e = \frac{n_i \mu_e}{e \chi_e}, \quad (3)$$

and similarly for holes. The photoexcited electrons and holes must diffuse together, giving ambipolar diffusion

$$D_a = (\sigma_e D_b + \sigma_b D_e) / (\sigma_e + \sigma_b),$$

from which

$$\frac{1}{D_a} = \left( e^2 \rho + e \frac{n_i}{n_i \mu_e} \right) \chi_e. \quad (4)$$

Here we have used $n_{ex} \ll n_0$. Our measurements suffice to specify neither $\chi_e$ nor $\mu_e$, but do constrain the relation between them: Eq. (4) shows that the larger the mobility, the larger $\chi_e$.

To further explore the relationship between $\mu_e$ and $\chi_e$, it will be instructive to recast it in terms of $m^*$ and the momentum-relaxation time $t_p$, and to compare our results on (Ga,Mn)As with the known properties of GaAs (which has $m^*/m_e = 0.063$). In undoped GaAs the mobility at 80 K can reach $2 \times 10^5$ cm$^2$/V s, but for $n \approx n_{ex}$ the mobility should be about $10^3$ to $10^4$ cm$^2$/V s, and comparable at 15 K.

The latter values correspond to a momentum relaxation times $t_p$ of 0.04 to 0.4 ps. The range of typical GaAs conduction-band parameters appears as a thick, vertical lines in Figs. 3(a) and 3(b), where we choose to plot $t_p$ rather than $\mu_e$ since mobility contains both $t_p$ and $m^*$.

Turning back to our (Ga,Mn)As results, at finite temperature $\chi_e(m^*)$ must be calculated numerically. For this purpose we introduce a simple model that treats both bands as isotropic and parabolic, the hole mass as 0.53$m_e$, and the conduction-band effective mass $m^*$ as a free parameter. We calculate first $\chi_e(m^*)$, then the corresponding $\mu_e(\chi_e, D_a, \rho)$, and finally the momentum-relaxation time $t_p = \mu_e m^*/e$; the results for 80 K appear in Fig. 3(a). Since $n_i$ is unknown, we repeat the calculation for a range of $n_i$: The solid curve assumes $n_i/n_{ex} = 0.1$, and we shade the entire possible region $0 < n_i/n_{ex} < 1$. Note that all points on the diagonal line for $n_i = 0$ have $\mu_e = 7700$ cm$^2$/V s, and that points in the allowed region above the line have yet higher mobility. We believe, therefore, that $\mu_e > 7700$ cm$^2$/V s; this value is high, but not without precedent in GaAs.

Figure 3(b) shows the results of a similar calculation for our 15-K data. For the region above the diagonal, $\mu_e \gtrsim 12,000$ cm$^2$/V s. The calculation assumes spin-splitting of the conduction band; a calculation without spin splitting would give the same lower bound on $\mu_e$, but would shift the curved boundary (corresponding to $n_i = n_{ex}$) leftward. Within the experimental uncertainties, the 15-K mobility is similar to that at 80 K.

It may seem that (Ga,Mn)As, as a disordered material, ought to have mobility well below that of GaAs. Indeed, our experimental results do not, strictly, exclude a low mobility: It is allowed if $m^*/m_e < 0.03$ (Fig. 3(a), lower left). We are not, however, aware of any mechanism to suppress the conduction-band density of states so strongly. It would also seem unlikely for $t_p$ to greatly exceed 0.4 ps, which suggests that $m^*$ cannot much exceed the GaAs value. Thus, the most plausible scenario supported by our data is that neither the conduction band’s scattering rate nor its effective mass is much influenced by Mn doping. We note that some recent results indicated that the (Ga,Mn)As valence band has GaAs-like effective mass and a low scattering rate. Indeed, those results are even more surprising than ours, because Mn states are much closer to the valence band edge than to the conduction band.

The high diffusivity we measure could aid the operation of a GaMnAs-based magnetic bipolar transistor. In order to exhibit high current gain, the device needs a minority-carrier diffusion length that greatly exceeds the base width $w$. The electrons’ low-temperature diffusion length of $L = \sqrt{D_e t_0} \approx 170$ nm is not long, but is long enough to allow $w < L/8$ to be achieved by commercially available photolithographic techniques. If the electrons’ spin diffusion $D_s$ is close to $D_e$, such a device would inject a spin polarization proportional to $D_s$ into the collector. Just as our measurement photoexcites enough electrons to fill the traps, the transistor would need to operate in a high-injection regime. High injection mitigates carrier trapping, and may also be responsible for the electrons’ high mobility: An $As^+$ antisite becomes neutral when trapping an electron.

In conclusion, we performed transient-grating experiments to measure the diffusion of photoexcited electrons in Ga$_{94}$Mn$_{8}As$. We found a signal consisting of three components; the component revealing motion of free electrons had a diffusivity of 60–80 cm$^2$/s at 80 K and 50–70 cm$^2$/s at 15 K. This rather high diffusivity, along with the measured
resistivity, constrains the electrons to have either a high mobility or a low effective mass. We consider it most plausible that the electrons’ effective mass and mobility are both comparable to those of GaAs, with $\mu_e \geq 7700 \text{ cm}^2/\text{V s}$. The mobility is nonetheless surprisingly high, given the disorder in (Ga,Mn)As alloys, and reflects favorably on the possibility of comparable to those of GaAs, with $\mu_e/C_0$ of $v_e = 0$ exceeds $10^{19} \text{ cm}^3/\text{V s}$ at 15–35 K and 80–100 K; the range of temperatures reflects likely heating of electrons.

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21In calculations, we incorporate this heating by using electron temperature of 35 K (100 K) for measurements at lattice temperature of 15 K (80 K).
22Diffusivity varies with $n_{so}$ and the peaks of our grating will diffuse more rapidly than the troughs. Our numerical simulations indicate that for the densities and temperature used in this experiment, this effect can cause us to underestimate $D_0$ by up to 30%, making electron mobility potentially somewhat higher, but not lower, than the values we report.
38$\mu_e/C_0 \approx 10–11.5 \text{ m}^2/\text{V s}$ at $15–35 \text{ K}$ and $80–100 \text{ K}$; the range of temperatures reflects likely heating of electrons.
39$\mu_e/C_0$ depends on $p_0$ and the hole density of states, neither of which is accurately known. However, the effect of the holes on $\mu$ is less than 8% provided $p_0$ exceeds $10^{18} \text{ cm}^3/\text{V s}$ and the hole effective mass exceeds 1/20 of the free-electron mass.