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Picosecond imaging of photoexcited carriers in quantum wells: Anomalous lateral confinement at high densities

L. M. Smith,* D. R. Wake, and J. P. Wolfe
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

D. Levi and M. V. Klein
Department of Physics, Materials Research Laboratory and Coordinated Science Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

J. Klem, T. Henderson, and H. Morkoç
Coordinated Science Laboratory, University Illinois at Urbana-Champaign, Urbana, Illinois 61801
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We have measured the in-plane motion of photoexcited carriers in semiconductor quantum wells and have discovered several surprising results. The effective diffusivity of the carriers at densities below $n = 2\times10^{11}$ cm^{-2} is found to depend upon the excitation level, possibly indicating defect-limited diffusion or phonon-wind effects. Above this density the spatial profiles exhibit two distinct components with widely differing diffusivities. We postulate that the slowly diffusing component represents carriers which are "thermally confined" to a phonon hot spot.

The existence of high-quality semiconductor quantum wells has provoked numerous studies of the kinetics of carriers confined in these structures. The physics of two-dimensional (2D) electronic transport involves the basic interactions of electrons with phonons, defects, impurities, and interfaces. An important question is whether the carrier transport in a quantum-well structure differs significantly from that in bulk crystals.[1] Specifically, one would like to know the 2D diffusivity of carriers (electrons and holes) and their mobility in applied fields. Both electrical and optical techniques have been employed to study the carrier motion within the confining plane of the quantum well.[2] Generally, the semiconductor is selectively doped so that one type of carrier (electron or hole) dominates the transport.

At low temperatures, photoexcited electron-hole pairs can bind to form excitons. The diffusion of these neutral particles in a quantum well has been observed by Hegarty and Sturge[3] using an optical transient grating technique. Significant questions remain about the scattering processes and the possible localization of the excitions due to fluctuations in the well potential. Also, at sufficient excitation levels, electron-hole plasma is produced, and very rapid expansion velocities of this phase have been inferred from time-resolved light scattering experiments.[4]

In the present Rapid Communication, we report direct measurements of ambipolar (electron-hole pair) transport in an undoped quantum well by combining picosecond laser pulses with optical imaging techniques. Following excitation by an intense laser pulse, the spatial distribution of electron-hole pairs is observed by measuring the optical transmission of a delayed probe pulse. Such a measurement directly yields the effective diffusivity of the photoexcited carriers. We have discovered that the motion of the photoexcited carriers in the plane of the quantum well depends markedly on excitation level and that at high carrier densities two spatially distinct components are observed with widely different diffusivities. We discuss possible interpretations of these results.

The sample used in our experiments is a [001]-oriented $GaAs/Al_{0.3}Ga_{0.7}As$ multiple quantum well (MQW) with 30 periods of 210-Å wells and 100-Å barriers grown by molecular-beam epitaxy. A 1-mm hole was etched through the substrate to allow absorption measurements. The sample is cooled by a constant flow of cold He gas, and the temperature is monitored by a calibrated carbon resistor.

Our experiments are a variation of the standard pump-probe method. A mode-locked argon-ion laser synchronously pumps two dye lasers. The intense 1-ps pump pulse is tuned to a wavelength (720 nm) which is about five LO phonon energies above the lowest-lying subband, as indicated in Fig. 1(a). A ten-times weaker 8-ps probe pulse is tuned resonantly to the peak of the heavy-hole exciton absorption line (at approximately 815 nm). After delaying the probe pulse relative to the pump pulse, these beams are separately focused to about 3-μm spots onto the crystal. The position of the probe beam is scanned as indicated in Fig. 1(b). Without the pump beam, the probe light is partially absorbed as it resonantly creates excitons in the quantum well. This excitonic absorption decreases (i.e., "bleaches") when a high density of carriers is present.[5-7] The change in the transmitted intensity of the probe light is approximately proportional to the local pair density, for the densities involved in our experiments.[5,7]

Figure 2(a) shows the spatial evolution of photoexcited carriers following absorption of a pump pulse with 0.1-pJ energy. The initial distribution of carriers has a FWHM of 4 μm corresponding to an average density[8] of approxi-

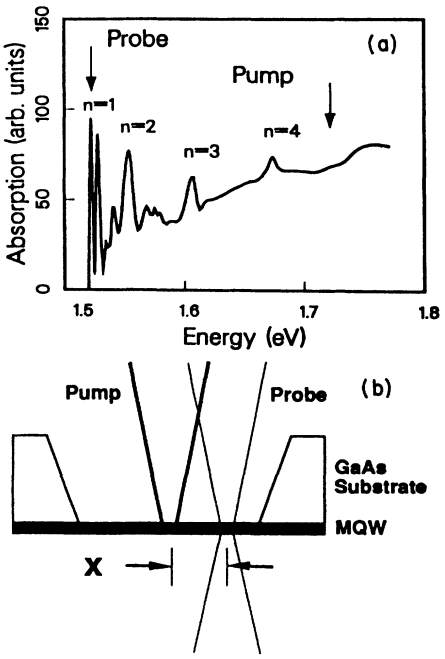

FIG. 1. (a) Absorption spectrum (Ref. 19) of 210-Å GaAs MQW. Allowed excitonic optical transitions are labeled by their principal quantum number. The positions of pump and probe photon energies are shown by the arrows. (b) Block diagram of sample showing relative positions of pump and probe beams.

mately 1×10^{11} cm^{-2}. The bath temperature is 8.5 K. Significant expansion of the photoexcited carrier distribution is observed in the first 1000 ps. Notably, this method is capable of measuring lateral expansion of carriers in the well for several nanoseconds after the pump pulse. During the first 4 ns after excitation, the carrier distribution expands laterally to about 20 μm, corresponding to an increase in area of 25 times that of the initial laser spot.

If the spatial expansion of these carriers is truly diffusive, their density $n(\mathbf{r},t)$ should obey the diffusion equation

$$\frac{\partial n}{\partial t} = -\frac{n}{\tau_l} + D\nabla^2 n , \tag{1}$$

where, for simplicity, the diffusion constant D and lifetime τ_l of the photoexcited carriers are assumed to be independent of carrier density. The solution to this equation is

$$n(r,t) = \frac{1}{4\pi Dt} e^{-r^2/4Dt} e^{-t/\tau_l} . \tag{2}$$

From this equation one can see the advantage of collecting spatial profiles at selected time intervals. The lifetime of the carriers affects only the overall intensity of the distribution, and the time dependence of the full width at half maximum, Δ, is given by

$$\Delta^2(t) = 11.08Dt + \Delta_0^2 . \tag{3}$$

The solid curves in Fig. 2(a) are simple Gaussian functions fit to the data. The square of their full widths at half maximum, Δ, vary linearly with time and yield an experimental diffusivity $D_{\text{eff}} = 74$ cm^2/s. We denote this as an effective diffusivity for reasons apparent below.

A remarkable discovery is that D_{eff} as well as the *shape* of the spatial profiles depend on excitation level. For example, at roughly three times lower excitation power than that of Fig. 2(a), D_{eff} drops by about a factor of 3. In addition, at higher powers, the spatial profiles take on a completely different character. Figure 2(b) shows that at an initial density of 3×10^{11} cm^{-2}, the spatial profile cannot be fit by a single Gaussian curve. A sharp structure now appears in the data. We have characterized these distributions as the sum of two Gaussian components, as indicated by the dashed lines in Fig. 2(b). This sum (solid line) describes the data quite well. A plot of Δ^2 for each of these components, as shown in Fig. 3(a), indicates their widely different diffusivities.

As the laser power is further increased, the slowly diffusing component begins to dominate the distribution, so that by $n_0 = 1\times10^{12}$ cm^{-2} the data are adequately described by a single, slowly expanding Gaussian distribution [Fig. 2(c)]. This unusual behavior is summarized in Fig. 3(b), which displays the three distinct regimes described above.

To interpret this complex behavior we now consider separately the interactions of the carriers with defects, other carriers and phonons. The power dependence of D_{eff} at low excitation levels could be explained by a distribution of trapping centers (e.g., associated with impurities or well-width fluctuations) which become saturated as the carrier density is raised. Localization of resonantly created excitons was previously indicated in the transient grating experiments of Hegarty and Sturge.[3] What, then, is the origin of the slowly diffusing component at *high* densities? One interesting possibility is the formation of a two-dimensional electron-hole liquid (2D EHL), produced by the attractive many-body interactions of electrons and holes. The possibility of a 2D EHL has been considered theoretically,[9-11] but never observed experimentally. While this is an intriguing interpretation, we feel that gas-liquid condensation is unlikely at the rela-

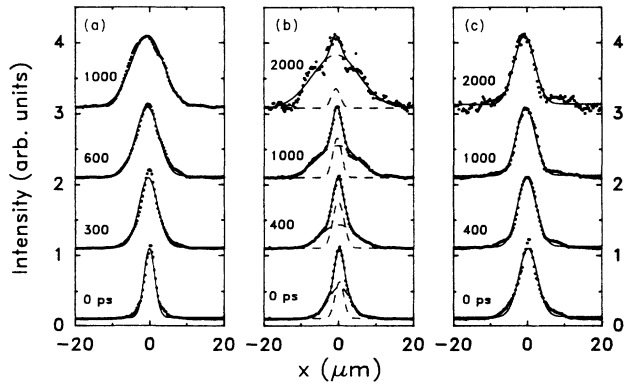

FIG. 2. Time-resolved spatial scans of carrier distributions after photoexcitation by pump pulse. (a) $n_0 = 1\times10^{11}$ cm^{-2}. (b) $n_0 = 3\times10^{11}$ cm^{-2}. (c) $n_0 = 1\times10^{12}$ cm^{-2}. Solid lines are best fits to the data using one or two Gaussian components. Those distributions where two components are used display each component as dashed lines.

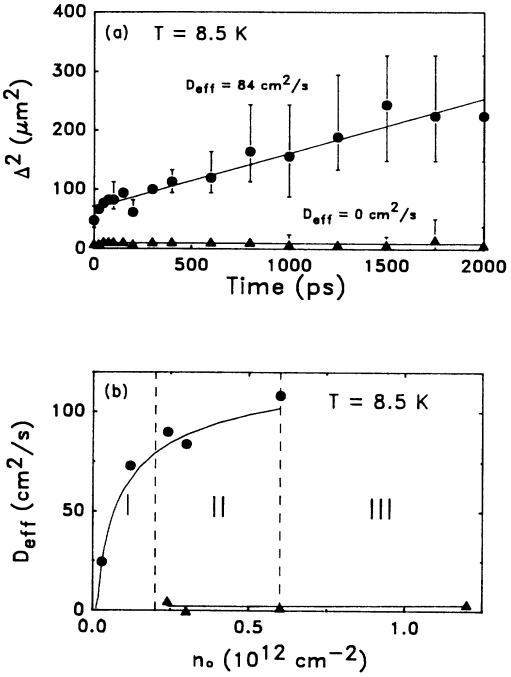

FIG. 3. (a) Plot of Δ^2 from fits in Fig. 2(b) ($n_0 = 3\times10^{11}$ cm^{-2}). Circles (triangles) are Δ^2 of broad (narrow) distribution. Straight lines are best fits to the data, with slopes proportional to D_{eff} as described in the text. (b) Plot of D_{eff} with increasing power. Circles (triangles) correspond to broad (narrow) distributions.

tively "high" experimental lattice temperatures of 8.5 K.

Another, quite different interpretation of the data is possible. We note that in these experiments the photoexcited carriers are created with a significant excess energy which is rapidly dissipated into nonequilibrium phonons. In bulk materials, it is known that such nonequilibrium phonons can greatly affect the transport of the photoexcited carriers [e.g., phonon-wind forces on EHL and EHP in Si and Ge (Refs. 12–14)]. Thus, a phonon-wind force increases with excitation power and thus leads to power-dependent expansion velocities, such as those observed in region I of our experiments [Fig. 2(b)]. The saturation of D_{eff} in region II implies a limit to the drift velocities which may also be understood within the context of a phonon-wind force. At this intermediate excitation level, the initial expansion velocity of the rapidly moving component is close to the transverse sound velocity of acoustic phonons, which is the maximum velocity that these phonons can drive the carriers. This model assumes that the diffusivity of the carriers is quite low (< 25 cm^2/s at 8.5 K), and that the net phonon flux determines the observed carrier drift velocity.

At this stage it is difficult to predict the form of the expansion width $\Delta(t)$ due to a phonon-wind force, which is a *drift* process. For a three-dimensional expansion of EHL in Ge, $\Delta^3 \propto t$ was predicted from phonon-wind theory[12] and verified experimentally.[15] Similar arguments predict $\Delta^2 \propto t$ for phonon-wind driven transport in two dimensions.[16] This dependence is indistinguishable from a *diffusive* process [Eq. (3)]. Further experimentation could differentiate between these two processes by changing the excess energy provided by the pump laser.

The existence of a high density of nonequilibrium phonons also suggests a novel explanation for the slowly diffusing component in regions II and III. For high phonon densities a region of diffusive phonon transport —known as a hot spot[13,17,18]— may be formed. Carriers in this region would not experience the net force associated with ballistic phonons and thus would be "thermally confined." The narrow component observed in the carrier distribution would define the spatial extent of the diffusive phonons, while those carriers outside of this region would be driven by the ballistic phonons escaping from the surface of the hot spot. This would explain the two-component behavior we observe. A reduced band gap in the heated region would also lead to thermal confinement; however, time-resolved absorption shows only a small spectral shift (< 1 meV) in the present case.

In summary, we have directly observed the macroscopic lateral motion of photoexcited carriers in a quantum-well structure. We believe that the carrier state is excitonic at the lowest densities ($n \leq 3\times10^{10}$ cm^{-2}) and EHP at the highest densities ($n \geq 3\times10^{11}$ cm^{-2}). No evidence is found for the highly supersonic carrier transport inferred from Raman-scattering experiments.[4] Instead, we have discovered that the carrier distribution displays two distinct spatial components, each with a surprising behavior. The faster moving component, which is dominant at *low* densities, exhibits an effective diffusivity which is power dependent, suggesting a phonon drift process. The slower moving component, which dominates at *high* excitation powers, is postulated to arise from the thermal confinement of carriers within a phonon hot spot. These unanticipated results reveal a variety of interesting physical processes in the dynamics of nonequilibrium carriers in semiconductor quantum wells.

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*Present address: IBM Thomas J. Watson Research Center, P. O. Box 218, Yorktown Heights, NY 10598.

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