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Picosecond time-resolved Raman studies of the expansion of electron-hole plasma in GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well structures

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The spatial expansion of the photoexcited electron-hole plasma created by picosecond-pulse laser excitation in GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well structures is investigated by time-resolved Raman scattering on a picosecond time scale. The experimental results show that the measured drift velocity of the plasma is comparable to its Fermi velocity. Consequently, our measurements demonstrate that the transport of the photoexcited electron-hole plasma in GaAs-$Al_xGa_{1-x}As$ quantum wells is thermodiffusive.

I. INTRODUCTION

The evolution of the electronic and thermal energy following the creation of the photoexcited electron-hole plasma (EHP) in semiconductors is a subject of great interest and controversy. From the theoretical point of view, Wautelet and Van Vechten[1] have inferred that the plasma generated by nanosecond laser pulses will be confined near the excitation spot. On the other hand, by fitting the luminescence data, Forchel, Schweitzer, and Mahler[2] have suggested that the plasma diffuses rapidly into the crystal at a velocity comparable to its Fermi velocity. Recently, Steranka and Wolfe[3] have performed time-resolved luminescence imaging on the EHP created by nanosecond-pulse laser excitation in Si. This spatial imaging experiment has indicated that (1) for low-intensity excitations, the plasma expands from the excitation spot at subsonic velocities, and (2) for very-high-intensity excitations, the plasma expands as a shell at near-sonic velocities and the plasma is actually driven by the phonon wind. These measurements, which are in contrast to the fast-diffusing model of Forchel *et al.*,[2] demonstrate unambiguously that on a time scale of the order of a nanosecond after the generation of the plasma, the plasma expands at subsonic or near-sonic velocities. However, we note that the spectroscopic analysis made by Forchel *et al.*[2] involves a time scale much shorter than a nanosecond; therefore, in order to avoid possible averaging effects and to resolve this discrepancy, a spatial imaging technique that has a time resolution of the order of a picosecond or less has to be employed. The expansion of the EHP has been studied on the picosecond time scale in bulk GaAs (Refs. 4 and 5) and in bulk CdSe (Ref. 6). These studies seem to support the fast-diffusion model of Forchel *et al.*[2] We note that all of the measurements reported so far have been obtained under a three-dimensional configuration. In order to investigate (1) what influence two-dimensional confinement and the zone-folding effect (referred to phonons) might have on the transport properties of EHP in a semiconductor, and (2) the evolution of the electronic and thermal energy following the creation of the photoexcited EHP in a semiconductor of reduced dimensionality, we have examined the spatial expansion of the photoexcited EHP in GaAs-$Al_xGa_{1-x}As$ multiple-quantum-well (MQW) structures by using time-resolved Raman scattering on a picosecond time scale. The local density and expansion velocity of the photoexcited EHP in the excited region of the GaAs quantum wells are obtained. The drift velocity of the plasma was found to be comparable to its Fermi velocity. Consequently, these measurements show that at low-intensity excitations the effects of reduced dimensionality and zone folding (referred to phonons) can be ignored and thermodiffusion governs the transport of the photoexcited EHP in GaAs-$Al_xGa_{1-x}As$ MQW's.

II. EXPERIMENTAL TECHNIQUE AND SAMPLE

The experimental setup is shown in Fig. 1. The excitation source is a DCM dye laser which is synchronously pumped by the second harmonic of an actively mode-locked cw yttrium aluminum garnet (YAlG) laser. It generates a pulse train of 3-ps light pulses at a repetition rate of 76 MHz. These pulses are split into two beams of different intensity (with 10:1 ratio) and orthogonal polarizations. The more intense beam is used to pump the photoexcited plasma and the other one probes these nonequilibrium excitations by spontaneous intersubband Raman scattering. The photon energy from the dye laser is chosen to be 1.870 eV; consequently, we would expect that the photoexcited carriers come mainly from excitations of the electrons from (a) the heavy- and light-hole subbands to the continuum states, and (b) the split-off hole subband to the first or second electronic subbands. However, because we do not observe any appreciable intersubband contribution from second or higher subbands to their next higher subbands in our Raman spectra taken under the excitations of (1) the probe pulse alone, and of (2) the pump and the probe pulses with different time delay Δt, we conclude that the photoexcited carriers primarily populate the first electronic subband for the parameters used in our experiments. The pump pulse is variably delayed before being

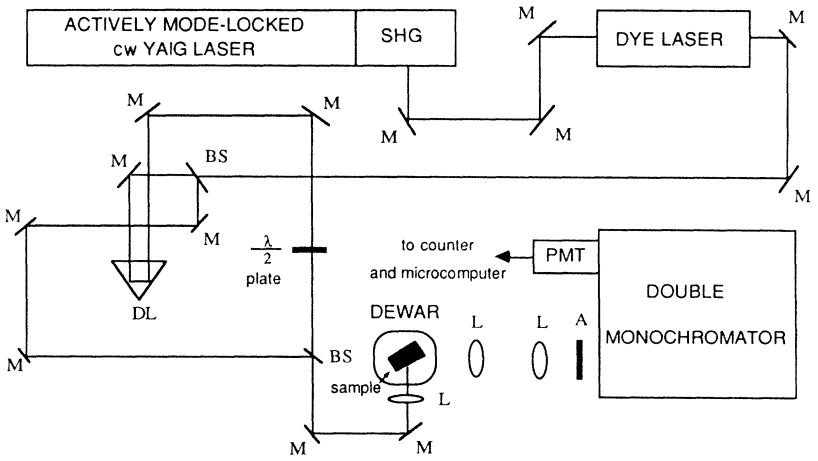

FIG. 1. Experimental setup for observing the spatial expansion of the photoexcited electron-hole plasma in GaAs-$Al_xGa_{1-x}As$ MQW's. SHG—second-harmonic-generation system, M—mirror, BS—beam splitter, DL—delay line, L—lens, A—analyzer, PMT—photomultiplier tube.

recombined with the probe pulse and focused to the surface of the sample. The carrier density generated by the pump pulse was about 4×10^{10} cm^{-2}. Raman scattering experiments were carried out in a backscattering geometry and in a pump-probe configuration. The undoped GaAs-$Al_xGa_{1-x}As$ MQW structure studied in this work was grown by molecular-beam epitaxy on a (001)-oriented undoped GaAs substrate. It consists of ~30 periods of 100-Å-thick $Al_xGa_{1-x}As$ ($x=0.3$) and 300-Å-thick GaAs layers. The samples were kept in contact with cold (~10 K) He gas.

The intersubband Raman scattering signal from photoexcited electrons confined inside the MQW's is made up of two parts which can be separated by a proper selection rule.[7] The first part consists of light scattering from spin-density fluctuations or single-particle excitations; the peak positions in the spectra reflect roughly (aside from the final-state interactions) the difference in subband energies which are involved in the transition. The second part comes from the scattering from charge-density fluctuations or collective intersubband excitations; because of the depolarization effect and coupling to LO phonons, its peak positions will shift in proportion to the carrier concentration. As shown in Fig. 2, for carrier concentration $n\leq1\times10^{11}$ cm^{-2}, this shift has been found[8,9] to be linear in n and about 1.55 cm^{-1} per 10^{10} cm^{-2} for our experimental situation. The shift of the peak position associated with the scattering of light from collective intersubband excitations was therefore utilized to determine the concentration of the photoexcited carriers in our measurements.

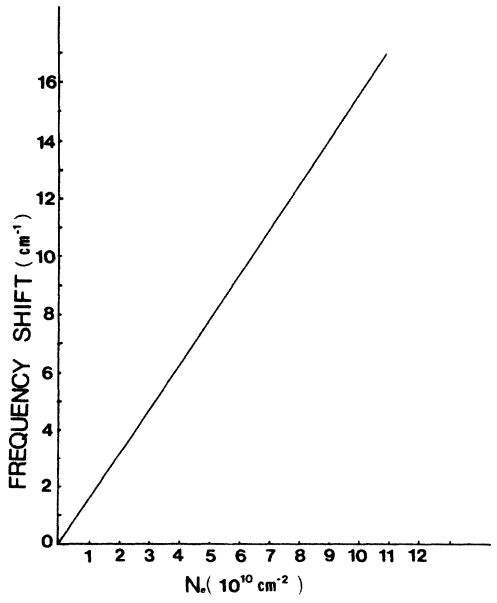

FIG. 2. Theoretical prediction of the frequency shift as a function of the electron density for collective intersubband excitations [from Eqs. (1) and (2) of Ref. 9].

In our experiment, the analyzer was placed along the [011] crystal direction, the pump and the probe beams were polarized along the [0$\bar{1}$1] and [011] directions, respectively. According to the selection rule, the probe beam will scatter collective intersubband excitations and the pump beam will have a contribution coming from single-particle excitations. However, due to the weakness of the latter at $n=4\times10^{10}$ cm^{-2} as well as the response of our detecting system, only the signal from collective intersubband excitations is observed in the spectra. The Raman signal is dectected by a computer-controlled Raman system.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

In Fig. 3, we have plotted the concentration of the photoexcited carriers as a function of the time delay Δt between the pump and the probe pulses for two samples: (a) a GaAs-$Al_xGa_{1-x}As$ MQW structure with 300-Å-thick GaAs layers, and (b) a GaAs-$Al_xGa_{1-x}As$ MQW structure with 300-Å-thick GaAs layers, but the dimension of the sample has been tailored to $\simeq$500 μm by chemical etching. The laser beams were focused to a spot of diame-

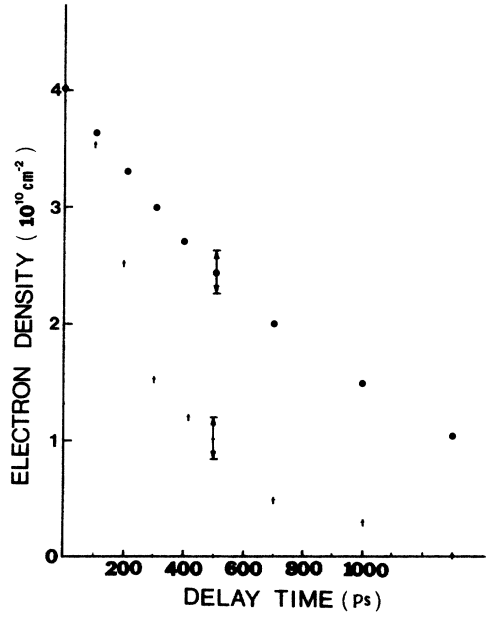

FIG. 3. The electron density vs the time delay Δt between the probe and the pump pulses. The filled circles are experimental data for GaAs-$Al_xGa_{1-x}As$ MQW's which has a diameter of $\simeq$500 μm and the spot size is $\simeq$500 μm. The crosses are for GaAs-$Al_xGa_{1-x}As$ MQW's, and the spot size is $\simeq$100 μm.

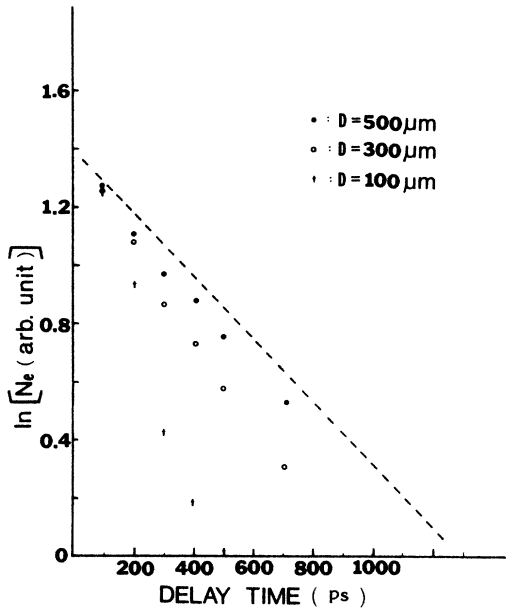

FIG. 4. The logarithm of the electron density vs the time delay Δt for three different laser spot sizes D = 500, 300, and 100 μm in GaAs-$Al_xGa_{1-x}As$ MQW's, although the spot size changes, the average power density in each case is kept the same. The dashed line (which corresponds to the experimental data for GaAs-$Al_xGa_{1-x}As$ MQW's having a diameter of $\simeq$500 μm and a spot size of $\simeq$500 μm) is also plotted for comparison. See text for discussion.

ter $\simeq$100 and $\simeq$500 μm on samples (a) and (b), respectively. We observe that the rate of decrease in the carrier concentration is much larger for sample (a) than for sample (b). Because the existence of the deep impurities in the GaAs sample has been shown to decrease the lifetime of the carriers,[10] one possible explanation is that sample (a) contains more deep impurities than sample (b). However, the following experimental observation rules out this possibility. In Fig. 4, the logarithm of the concentration of the photoexcited carriers is plotted against the time delay for sample (a) for three different laser spot sizes. We have deliberately kept the power density the same although we varied the spot size. These measurements show that the rate of decrease of the carrier concentration becomes smaller as the size of the focused spot increases, and eventually it approaches the value given in sample (b) of Fig. 3, which is indicated by the dashed line in Fig. 4. If deep impurities in the GaAs quantum wells are responsible for the observed data as shown in Fig. 3, we would expect that the rate of decrease in the carrier concentration should be independent of the size of the focused spot. Therefore, in order to explain our measurements, some other intrinsic dissipation mechanism has to be invoked. We attribute the observed phenomena to the expansion of EHP. Qualitatively, the experimental data in Fig. 4 can be realized as follows: If we start with the same excitation power density and, consequently, the same electron-hole concentration in the plasma, then because the initial velocity of the expanding plasma (which is comparable to its Fermi velocity) is the same, the lowering of the electron-hole pair density is more efficient for a smaller excited spot. Quantitatively, the observed data can be fully understood, provided that the plasma is expanding at a velocity of about 8×10^6 cm/sec. This measured plasma drift velocity is comparable to its Fermi velocity (about 8.6×10^6 cm/sec for $n=4\times10^{10}$ cm^{-2}). Consequently, our experimental results demonstrate that at low intensity excitations the transport of EHP in GaAs-$Al_xGa_{1-x}As$ MQW's is thermodiffusive and is consistent with the fast-diffusing model of Forchel *et al.*[2] We have tried to make the measurements at higher laser power density, but at these high-level excitations, the background signal (presumably resulting from the radiative recombination of hot electrons and holes excited through nonlinear processes) becomes so large that we cannot confidently analyze our data.

When we compare the experimental data of Collins and Yu[4] in bulk GaAs with our current results in GaAs quantum wells, we find out that the reduced dimensionality and zone-folding effect (referred to phonons) have little (if any) influence on the transport properties of EHP in a semiconductor such as GaAs. The transport properties of EHP in a semiconductor of direct band gap tend to be thermodiffusive; whereas those of indirect band gap follow the phonon-wind model. It is then appropriate to say that the dominant factor which determines the transport properties of the EHP in a semiconductor is the nature of its band gap. The reason why this is the case is not known, and more theoretical and experimental work are needed.

We have also performed similar measurements on GaAs-$Al_xGa_{1-x}As$ MQW's with different thicknesses of GaAs layers ($L_z\leq400$ Å). These experimental results show that the thickness of GaAs layers has very little effect on the transport properties of the EHP in GaAs-

$Al_xGa_{1-x}As$ MQW's.

In conclusion, we have used picosecond time-resolved Raman scattering to investigate the spatial expansion of photoexcited EHP in GaAs-$Al_xGa_{1-x}As$ MQW structures. The experimental results show that the measured drift velocity of the plasma is comparable to its Fermi velocity, and therefore demonstrate that the transport of the photoexcited EHP in GaAs-$Al_xGa_{1-x}As$ is thermodiffusive.

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