Light Scattering by Photoexcited Two-Dimensional Electron Plasma in GaAs-(AlGa)As Heterostructures

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We report resonant inelastic light scattering by photoexcited electrons in GaAs-(AlGa)As multiple quantum-well heterostructures and find that a variable-density two-dimensional electron-hole plasma is created in a controlled manner. The energy levels and collective electron-electron and electron-phonon interactions were studied for carrier densities as high as $\sim 3\times 10^{12}$ cm^{-2}. Charge neutrality is preserved in each quantum well. Complex collective interactions occur among modes involving transitions between quantum-well states.

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In this Letter we report the observation of resonant inelastic light scattering by photoexcited two-dimensional (2D) plasma in GaAs-(AlGa)As multiple-quantum-well (MQW) heterostructures. Single-particle and collective excitations are observed with selection rules as in modulation-doped MQW's.[1] This enables us to probe the energy level structure, collective carrier interactions, and coupling to longitudinal-optical (LO) phonons as a function of photoexcited carrier density.

Although stimulated emission[2] and laser oscillation[3] have been observed, light scattering from photoexcited 2D plasma has not been reported previously. Observations of light scattering from photoexcited 3D plasmas have been limited to either single-particle excitations in GaAs [4] and Si,[5] or collective excitations in GaP.[6] One of the puzzling aspects of the latter work was that collective mode energies were independent of photoexcitation intensity. In contrast, the energies of collective modes in our 2D system do change with photoexcitation intensity. This shows that a variable-density 2D plasma is created in a controlled manner, and that its properties can be studied by light scattering. We find that the quantum-well energy-level structure remains unchanged by photoexcitation for electron and hole densities as high as $\sim 3\times 10^{12}$ cm^{-2}. At high densities the spectra are rich in structure related to occupation of more than one quantum-well level and coupling between collective modes.

We studied several undoped GaAs-(AlGa)As MQW heterostructures grown by molecular-beam epitaxy on (001) GaAs substrates. Carrier photoexcitation was achieved with a cw Kr^+-ion laser operating simultaneously at 6471 and 6764 Å with a maximum power output of 450 mW. The laser beam was focused onto the sample, by means of a spatial filter and a high-quality lens, to a spot of radius ~ 10 μm. The absorption length is ~ 0.5 μm, or about ten superlattice periods. Photoexcited carriers are expected only in the GaAs layers. The carriers excited in the (AlGa)As layers are believed to either recombine quickly or become confined in GaAs wells. The density of carriers is anticipated to be largest in the GaAs layer at the sample surface and decrease for subsequent layers.

The 6471-Å line was used to obtain resonant inelastic light-scattering spectra. The photon energy, 1.916 eV, is close to the energy separation between the spin-orbit–split valence band and the lowest conduction band at the Γ point (the $E_0+\Delta_0$ optical gap). We measured polarized $z(x'x')\bar{z}$ and depolarized $z(y'x')\bar{z}$ backscattering spectra. z and $\bar{z}$ are the propagation directions of incident and scattered light, normal to the layers. The symbols in parentheses give the polarizations of the incident and scattered light. x' and y' are along (110) and (1$\bar{1}$0) directions in the plane of the layers. The samples were kept in contact with cold (~ 10 °K) He gas. Electron temperatures were estimated from Stokes–anti-Stokes intensity ratios. For the highest pump density ($P\approx 3\times 10^4$ W/cm^2), the electron temperature is ~ 70 °K.

Figure 1 shows spectra from a GaAs-($Al_{0.2}$-$Ga_{0.8}$)As MQW heterostructure. The $z(x'x')\bar{z}$ spectrum at a relatively low power density of $P\approx 200$ W/cm^2 shows a single peak at 36.6 meV

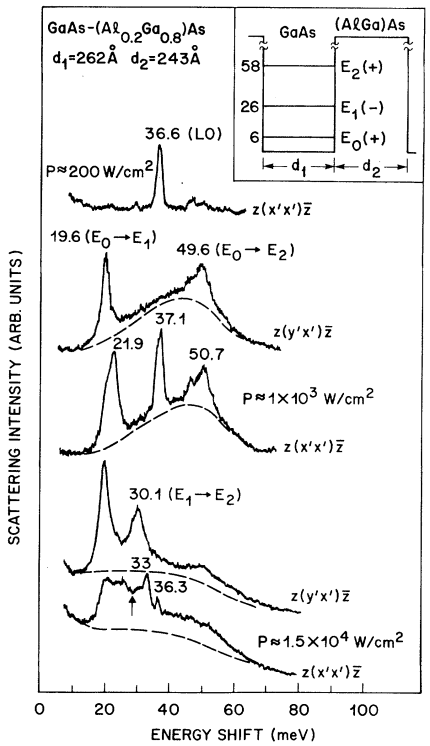

FIG. 1. Light-scattering spectra from a GaAs-$(Al_{0.2}Ga_{0.8})As$ MQW heterostructure for three different power densities. The dotted line indicates a background tentatively assigned to $E_0+\Delta_0$ hot luminescence. The inset shows the structure of the conduction-band edge and the calculated conduction-band quantum-well states in Ref. 8. d_1 and d_2 are the thicknesses of the GaAs and $(Al_{0.2}Ga_{0.8})As$ layers. The sample has 151 periods like the one shown.

(295 cm^{-1}), the LO phonon of GaAs. The LO_1 and LO_2 phonons of the $(Al_{0.2}Ga_{0.8})As$ layers[7] are too weak to be observed. At these power densities the $z(y'x')\bar{z}$ spectra show no identifiable structure because Raman scattering by optical phonons is forbidden. When the power density is increased to about 10^3 W/cm^2 new bands appear in both spectra. The new bands in $z(y'x')\bar{z}$ spectra remain at the same energies, 19.6 meV (158 cm^{-1}) and 49.6 meV (400 cm^{-1}), when the power density is increased. For higher power densities a new band appears at 30.1 meV (243 cm^{-1}) in $z(y'x')\bar{z}$ spectra. The position of this band is also independent of power density. New bands in $z(x'x')\bar{z}$ spectra have a markedly different behavior. When the power density is increased they shift to higher energies and broaden. At high power densities the $z(x'x')\bar{z}$ spectra are broad and rich in structure, as shown in Fig. 1 for $P\approx 1.5\times 10^4$ W/cm^2.

In order to determine the origin of the spectral bands that appear under high photoexcitation we calculated the energy levels in the GaAs wells.[8] The inset in Fig. 1 gives the energies and parities of the three lowest conduction quantum-well states. The $E_0\rightarrow E_1$ and $E_0\rightarrow E_2$ transitions are predicted at energies of 20 and 52 meV. These are very close to the energies of bands in $z(y'x')\bar{z}$ spectra with $P\approx 1\times 10^3$ W/cm^2. They are therefore assigned to the transitions of photoexcited electrons in the lowest quantum-well state. The additional band observed at 30.1 meV in $z(y'x')\bar{z}$ spectra with $P\approx 1.5\times 10^4$ W/cm^2 is assigned to the $E_1\rightarrow E_2$ transition. This shows that the density of photoexcited electrons in the GaAs wells increases with power density, and that for $P\approx 1.5\times 10^4$ W/cm^2 there is a substantial population of the first excited state. Since the bands in $z(y'x')\bar{z}$ spectra do not shift with changes in carrier density, we assign them to single-particle excitations. The features observed in $z(x'x')\bar{z}$ spectra, being strongly dependent on photoexcited carrier densities, are assigned to collective excitations.

We therefore find that light-scattering selection rules applicable here are identical to those for electrons in modulation-doped MQW heterostructures.[1,9] The $z(y'x')\bar{z}$ spectra correspond to spin-density intersubband excitations. These have single-particle character and are related to vertical transitions between the 2D subbands associated with conduction quantum-well states. The $z(x'x')\bar{z}$ spectra are due to charge-density collective intersubband excitations. Their energies are shifted by collective electron-electron interactions[1,10] and coupling to LO phonons.[1,9,11] Transitions by the photoexcited holes are not observed here, since their scattering intensities are not enhanced at the $E_0+\Delta_0$ optical gap. This result is consistent with the resonant mechanisms discussed in Ref. 9; light scattering by photoexcited holes cannot occur through intermediate virtual transitions from the spin-orbit–split-off valence band to the conduction band since the holes thermalize to the upper valence bands. With this understanding of the spectra we are now in a position to explore the energy-level structure and collective interactions as a function of photoexcited carrier density.

The energies and widths of bands in $z(y'x')\bar{z}$ spectra of single-particle transitions are found to be independent of power density, up to $P\approx 3\times 10^4$ W/cm^2. This indicates that creation of a high density of photoexcited electrons and holes does not result in a significant distortion of the confinement potential wells within the GaAs layers.

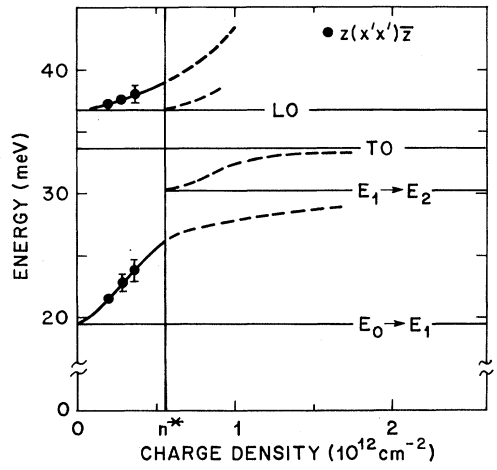

FIG. 2. Collective electron–LO-phonon modes of odd parity, associated with $E_0 \rightarrow E_1$ and $E_1 \rightarrow E_2$ transitions in sample 1. The full line is calculated according to Ref. 1. The coupling between the odd-parity modes, which occurs for $n > n^*$ when the first excited subband begins to be occupied, is illustrated by dashed curves. The experimental points for $n > n^*$ are shown as dots. The vertical bars indicate the widths of the spectral bands.

For this to occur, charge neutrality must be preserved in each layer. Consequently, we conclude that the photoexcited holes also become confined in the highest-energy quantum-well states associated with heavy and light valence bands.

In the interpretation of $z(x'x')\bar{z}$ spectra in terms of collective modes, we consider first the case of relatively low power density in which only the lowest subband is occupied. Figure 2 shows the calculated energies of the coupled collective modes of the odd-parity $E_0 \rightarrow E_1$ transition and an LO phonon as in Ref. 1, neglecting the effect of the photoexcited holes.[12] The bands observed at 21.9 and 37.1 meV for $P \approx 1 \times 10^3$ W/cm^2 (Fig. 1) are assigned to these collective modes. We show in Fig. 2 these experimental points placed at the charge density that gives the best agreement between the calculated and observed coupled mode energies. Experimental points obtained at two other excitation intensities are also shown in Fig. 2. The band observed at 50.7 meV in the $z(x'x')\bar{z}$ spectrum with $P \approx 1 \times 10^3$ W/cm^2 is assigned to the collective mode of the even-parity $E_0 \rightarrow E_2$ transition at 49.6 meV, and is not shown in Fig. 2.

For higher power densities we observe occupation of the first excited subband E_1. In this case the two odd-parity collective excitations, associated with $E_0 \rightarrow E_1$ and $E_1 \rightarrow E_2$ transitions, are expected to be coupled. We calculated an approximate value of their coupling Coulomb matrix element using envelope functions of the infinite square well, and found it to be close to the matrix elements of the uncoupled transitions.[1,11] In addition, these modes are also coupled to two LO phonons with depolarization electric fields of odd parity with respect to the reflection symmetry of the quantum well. We now expect four coupled modes as illustrated by dashed lines in Fig. 2.

We find evidence for such complex electron-electron and electron-phonon interactions in $z(x'x')\bar{z}$ spectra with $P \gtrsim 3 \times 10^3$ W/cm^2. The observed behavior is qualitatively consistent with the dashed curves in Fig. 2. However, the broad spectral line shapes prevent us from making a quantitative comparison at the present time. In the case of $P \approx 1.5 \times 10^4$ W/cm^2, shown in Fig. 1, the scattering between 22 and 33 meV is assigned to the two lowest coupled collective modes of odd parity. It appears as a quasicontinuum because different GaAs layers have different photoexcited electron densities. The dip in the scattering intensity indicated by an arrow just below the energy of the $E_1 \rightarrow E_2$ transition is explained by the noncrossing behavior of the coupled modes as shown in Fig. 2. The peak at 33 meV is the screened LO phonon which corresponds to the second branch in Fig. 2.

In the $z(x'x')\bar{z}$ spectrum with 1.5×10^4 W/cm^2 we also observe a sharp band at 36.3 meV, just below the LO-phonon energy at 36.6 meV. We assign this band to an LO phonon with even-parity depolarization field, which is coupled to the collective mode of the even-parity $E_0 \rightarrow E_2$ transition. It could not be identified in the $z(x'x')\bar{z}$ spectrum with $P \approx 10^3$ W/cm^2 because it was obscured under the band at 37.1 meV.

Results obtained from other samples are in general similar to those presented above. In the case of narrower ($d_1 = 150$–200 Å) quantum wells, in which quantized energy levels have a larger spacing, occupation of only one subband was observed over the whole range of power densities used in these experiments. We also found that the linewidths in the spectra of single-particle intersubband transitions vary in different samples. The dependence of the linewidth on the quality of GaAs-$(Al_{0.2}Ga_{0.8})$As interfaces is under investigation.

In summary, we have shown that in GaAs-(AlGa)As MQW heterostructures it is possible to create a variable-density photoexcited 2D electron-hole plasma in a controlled manner. The

energy-level structure, charge densities, and collective interactions can be studied and understood with inelastic light-scattering spectroscopy. Somewhat surprisingly, we find that the energy-level structure remains unchanged by photoexcitation, indicating that for densities as high as $\sim 3\times 10^{12}$ cm^{-2} (equivalent to $\sim 10^{18}$ cm^{-3}) there is no spatial separation between electrons and holes. Such measurements also provide a means to determine directly the electron energy-level structure in quantum wells. The collective excitations display a complex behavior due to coupling among different quantum-well excitations and also with LO phonons. The spectra presented here are broadened by the varying plasma density in different wells. Extension of these studies to single quantum wells should allow one to obtain more quantitative information from spectra.

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[12]Their effect on collective-mode energies is anticipated to be relatively minor because hole subband spacings and effective plasma energies are much smaller.

Electron Hopping Conduction in the Soliton Model of Polyacetylene

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It is shown that at low temperatures, phonon-assisted electron hopping between soliton bound states may be the dominant conduction process in a lightly doped, one-dimensional Peierls system such as polyacetylene. The presence of disorder, as represented by a spatially random distribution of charged dopant molecules, causes the hopping-conduction pathways to be essentially three dimensional. Calculated values of the conductivity, thermopower, and transverse spin diffusion constant compare favorably with experiment.

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Recently, considerable theoretical[1-4] and experimental[5-10] interest has been focused on the properties of *trans*-polyacetylene, $(CH)_x$. Probably the most successful theoretical model of $(CH)_x$ to date is the soliton model of Su, Schrieffer, and Heeger[1] (SSH), and others.[2] In this paper, certain effects of the presence of disorder on the properties of the soliton model are considered. The results provide a satisfactory explanation of the transport properties of lightly doped polyacetylene. In addition, some novel aspects of the conduction mechanism, electron hopping between dynamical defects, make the problem quite interesting in its own right. In a forthcoming paper (Ref. 3, hereafter called paper II) a microscopic model of polyacetylene is considered, and the present phenomenological results are made quantitative.

According to the model of SSH, undoped polyacetylene is a semiconductor due to a commensurate Peierls distortion which doubles the unit cell (dimerization) and opens a band gap at the Fermi

