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Plasmons and phonons bound to neutral donors in semiconductor quantum-well structures

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We report resonant Raman scattering studies of selectively doped GaAs/$Al_xGa_{1-x}As$ quantum-well structures. This system can be modeled as a quasi-two-dimensional hydrogenic donor immersed in a degenerate electron gas in a polar lattice. The spectra show new collective excitations which we identify as intersubband plasmons and LO phonons bound to the neutral donor by the dipole field associated with the intersubband donor transition. The data can be understood quantitatively in terms of a simple phenomenological model which employs an effective dielectric function.

Over the years there has been a large and continuous interest in collective excitations in semiconductors. In particular, plasmons as well as coupled plasmon-phonon modes were studied intensely starting in the 1960s in bulk semiconductors,[1,2] and over the last 10 years in the quasi-two-dimensional (2D) GaAs/$Al_xGa_{1-x}As$ structures.[3] In addition, there has been a parallel effort to understand the interaction of LO phonons with an electron bound to a shallow donor. This interaction gives rise to a bound (localized) phonon.[4-8] In the Fröhlich model the phonons are treated in terms of a continuous polarization field which can respond to the charge of the electron. If a donor transition is made resonant with the phonon energy a localized phonon can occur. Within this approximation and to the extent that the donor remains bound we can consider the response of a quasi-2D electron gas to a donor in a similar way. An intersubband plasmon interacts with the donor through its polarization field, and can become localized in the same sense as a phonon. When both plasmons and LO phonons are present and their energies are resonant they will couple; if they are also resonant with a donor transition the coupled modes can become localized.

This situation is easily realized in GaAs/$Al_xGa_{1-x}As$ quantum-well structures. The quantum well confines the electrons in one dimension, increasing their energies, and allowing both an intersubband donor transition and the intersubband plasmon to be shifted into resonance with the LO phonon. We report Raman spectra from such samples that reveal the ordinary extended plasmon-phonon modes as well as new peaks which we identify as coupled intersubband plasmons and phonons localized at donor impurities.

The molecular-beam-epitaxy (MBE-)grown structure consists of alternating layers (nominally 200 Å) of GaAs, the center third of which were Si doped at an estimated density of 1×10^{16} cm^{-3}, and $Al_{0.25}Ga_{0.75}As$ with the center third also doped with Si at an estimated 8×10^{16} cm^{-3}. The wafer was cleaved into pieces 5 mm on a side and was characterized by photoluminescence, photoreflectance, cyclotron resonance, and Raman scattering. The well width was found to vary across the 3-in. wafer from $L_z=182$–200 Å. The free-electron concentration per well was found from cyclotron resonance measurements to be $n=1.5\times10^{11}$ cm^{-2}. This value is consistent with the plasma shifts[3] of the free-electron transitions observed in the resonant Raman scattering (RRS) spectra. The electron concentration was reduced by a 2-MeV electron irradiation.[9] This procedure introduces deep levels into the sample which trap some of the electrons thereby reducing the Fermi level.

The RRS measurements were taken with the laser energy E_L in resonance with the confinement-shifted GaAs $E_0+\Delta_0$ and E_0 gaps.[3] The spectra were taken in the $z(*,*)\bar{z}$ backscattering geometries where z and $\bar{z}$ are the directions of the incident and scattered light, respectively, and $(*,*)$ designates the polarizations of the incident and scattered light ($*=x',y',x$, or $y\equiv[110],[1\bar{1}0],[100]$, [010], respectively). With these scattering geometries the spin-density (perpendicular polarizations) and charge-density (parallel polarizations) electronic excitations can be probed separately. In the spin-density spectrum, the Raman peaks from the electronic transitions approximately yield the single-particle (SP) transition energies.[3] In contrast, the peak positions measured in the charge-density spectrum are shifted from the SP transition energies as a consequence of interactions between the excitations via their electric fields.[3]

Spectra taken in two polarization configurations are shown in Fig. 1. The doublet in the $z(x',y')\bar{z}$ spectrum occurs at an energy which corresponds approximately to the E_0-E_1 intersubband transition energy which we calculate using the potential-well model including the static Hartree potential.[10] It has been shown previously[11,12] that the electron intersubband transition splits into a doublet when there are neutral donors present. These studies were performed on donors in a small concentration of photoexcited electrons.[11,12] The peaks in the doublet were identified in those works according to the SP energy diagram shown in the inset in Fig. 2. We agree with this identification except that, in our case, we find that the intersubband donor transition is the lower-energy part of

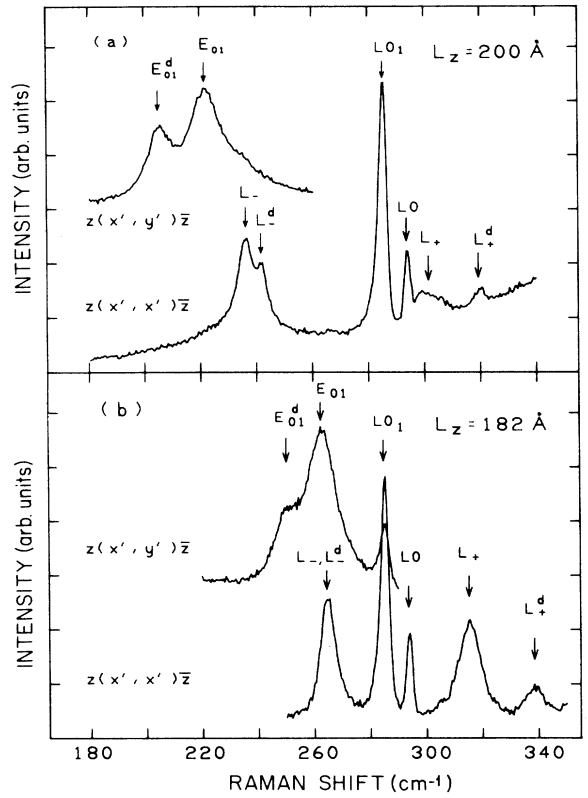

FIG. 1. Raman spectra obtained with two scattering geometries at $T=4$ K and $E_L=1.90$ eV on a sample with (a) $L_z=200$ Å and (b) $L_z=182$ Å. $n=1.5\times10^{11}$ cm^{-2}. The $z(x',y')\bar{z}$ and $z(x',x')\bar{z}$ spectra arise from SP and collective excitations, respectively.

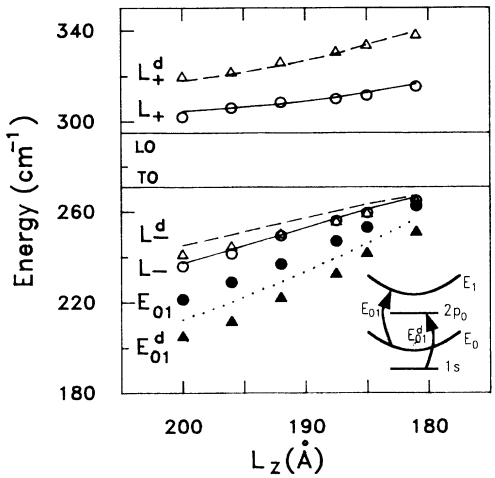

FIG. 2. Peak energies of SP (closed symbols) and collective (open symbols) excitations as a function of well width with $n=1.5\times10^{11}$ cm^{-2}. The solid and dashed lines are the results of calculations described in the text. The dotted line is the donorlike collective excitation which is predicted but not observed. The inset shows schematically the SP transition assignments.

the doublet. This identification is based on its temperature dependence. As the temperature is increased the lower-energy peak shifts into the higher-energy one. At 150 K only the E_{01} peak remains, consistent with the expectation that at high temperatures all the donors should be ionized. In addition, as the electron concentration is reduced the E_{01}^d peak shifts up in energy toward the E_{01} peak. This leads us to believe that the reduction in energy of the donor transition relative to E_{01} with the addition of the electron gas is due to screening effects. In this paper we are concerned with the collective excitations which are derived from SP transitions and will not consider the origin of this screening in any more detail.[13] A detailed study of the static screening of the donor will be published separately.

The $z(x',x')\bar{z}$ spectrum is also shown in Fig. 1. In addition to the LO_1 and LO peaks which occur at the GaAs-like LO phonon energy of bulk $Al_{0.25}Ga_{0.75}As$ and the LO phonon energy of bulk GaAs, there are four other peaks: $L-$,L_-^d and $L+$,L_+^d. Although the $L-$ L_-^d peaks are not resolved at $L_z=182$ Å, they clearly are at $L_z=200$ Å. This spectrum (ignoring the doublet structure for the moment) resembles the collective excitation spectra measured for a free quasi-2D electron gas with no donors. Such spectra are known to originate from the two coupled modes resulting from an intersubband plasmon interacting with the GaAs LO phonon through their polarization fields.[3] The energies of such modes can be calculated, and for this well width and electron concentration would occur very close to the energies of the $L-$ and $L+$ peaks. With the addition of neutral donors into the electron gas, we find that each peak splits into a doublet just as the SP spectrum did, and that it is the higher-energy parts of the doublets which are associated with the SP donor transition (i.e., which shift with temperature). We interpret these extra modes as due to coupled intersubband plasmons and phonons which are localized at the neutral donors.

The SP and collective excitation spectra were measured in each piece of the wafer and the energies of the peaks are plotted in Fig. 2. The well-width values L_z of these data were obtained by fitting E_{01} to a calculation of the SP subband energies.

The spectra of two of these samples ($L_z=187$ and 200 Å) were also measured as a function of electron concentration n and the peak energies of the $L_z=187$ Å sample are plotted in Fig. 3. Because L_+^d is better resolved at $L_z=187$ Å, we have chosen these data to illustrate the density dependence. As described above, n was reduced by high-energy electron irradiation. The calibration of n versus fluence was obtained by fitting $L+$; i.e., we used the result (discussed below) that the $L+$ mode is unperturbed by the donors and used the standard calculation for this energy as a function of density.[3] The gross aspects of the data which we consider especially significant are the positive-energy shifts of the L_-^d and L_+^d with respect to $L-$ and $L+$, respectively (in Fig. 2), and also the fact that the L_+^d peak does not shift back to the LO phonon energy as n approaches 0 (in Fig. 3).

To interpret the data we start with results obtained from previous studies on the interaction of a neutral donor with the LO phonon in a bulk semiconductor.[4-8] Micro-

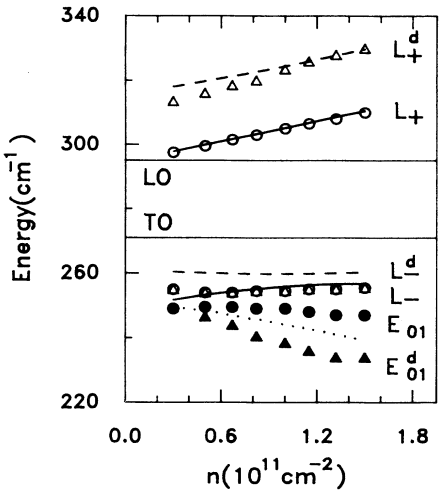

FIG. 3. Peak energies as a function of electron concentration for $L_z = 187$ Å.

scopic calculations using the Fröhlich interaction have shown that the phonon can become localized at the donor when a transition energy of the donor is nearly resonant with the phonon energy.[4,6–8] In addition, Barker was able to treat this problem in a phenomenological way with a macroscopic dielectric function technique.[5] Because the plasmon carries a polarization field which is similar to that of the LO phonon, it is a simple matter for us to extend his model and to include both the phonon and the plasmon.

In this approach an effective dielectric function for the medium, $\epsilon_{\text{eff}}(E)$ (where E is the energy), is derived, allowing the energies of the collective modes of the system to be found from $\epsilon_{\text{eff}}(E)=0$. To derive $\epsilon_{\text{eff}}(E)$ each donor is replaced with a dielectric sphere of radius a_s, which should be on the order of the Bohr radius. The dielectric function within the sphere differs from that of the background, ϵ_m, by the contribution from the donor's dipole moment:

$$\epsilon_s(E)=\epsilon_m(E)-\epsilon_\infty\frac{E_d^2}{E^2-E_{01}^{d2}},$$

where the oscillator strength of the dipole transition is written in terms of a squared energy E_d^2. The background dielectric function will have contributions from the phonon and the intersubband plasmon:[14]

$$\epsilon_m(E)=\epsilon_\infty\left[1-\frac{E_{\text{ph}}^2}{E^2-T^2}-\frac{E_p^2}{E^2-E_{01}^2}\right],$$

where $E_{\text{ph}}^2=L^2-T^2$, T=TO phonon energy, L=LO energy, and E_p is the plasma energy shift of the intersubband plasmon. The polarizability of a single sphere in the background dielectric medium is found, and the effective dielectric function is then derived for the two-dimensional gas of spheres.[15] The result is a Clausius-Mossotti-type equation with a filling factor (f) which is defined as the volume of the sphere times an effective 3D density of spheres:

$$\epsilon_{\text{eff}}(E)=\epsilon_m\left[1+\frac{3(\epsilon_s-\epsilon_m)f}{\epsilon_s+2\epsilon_m-(\epsilon_s-\epsilon_m)f}\right].$$

The collective excitation energies are given by the zeros of this equation and consist of two sets. One set, given by the two zeros of $\epsilon_m(E)$, contains just the coupled plasmon-phonon modes in the absence of donors. The other set contains the new localized modes L_-^d, L_+^d and a third, undetected mode which is the donorlike collective excitation. The only unknowns in these equations are the radius of the sphere and the oscillator strength of the donor. Choosing these to be the constants $a_s=140$ Å and $E_d^2=3E_p^2$ (where E_p is the value at $n=1.5\times10^{11}$ cm^{-2}), we obtain the fits shown in Figs. 2 and 3. The value of a_s is within a factor of 2 of the 3D Bohr radius (100 Å in GaAs). The oscillator strength of the donor transition depends on the microscopic details of the quasi-2D donor, which are not well understood. We can, however, make a rough estimate of the oscillator strength using the wave functions of the 3D donor, and we find that this estimate is within a factor of 2 of the value employed in Figs. 2 and 3.

Additional tests of the validity of the model are obtained by the comparison of the observed and predicted Raman scattering intensities. When the scattering occurs from the total polarization of the excitation, the cross section will be simply proportional to $\text{Im}(1/\epsilon_{\text{eff}})$.[3,16] The spectra we have measured at the $E_0+\Delta_0$ gap are not expected to be related to ϵ_{eff} in such a simple fashion because they include a large contribution from the charge-density fluctuation mechanism.[3] However, scattering in resonance with higher-lying subbands of the E_0 gap, which are unoccupied before or after the transition, occur mainly, we believe, via the forbidden Fröhlich mechanism[3] and therefore via the total polarization of the excitation. In fact, the spectra at the peak of the resonance enhancement near the third heavy-hole–electron energy gap compare very well with the function $\text{Im}(1/\epsilon_{\text{eff}})$. Furthermore, this calculation gives a donorlike collective excitation intensity much smaller than the other peaks, which is consistent with its absence in the data. Although we might anticipate that the scattering from the donorlike excitation could be enhanced to the point of detection by some as yet not understood resonance condition at another laser energy, we have not found this to be the case after considerable scanning of the laser energy.

The agreement of the measured spectra with the calculated spectra (both energies and intensities) using physically reasonable fitting parameters supports our interpretation of the data in terms of this model. With this interpretation we can understand most aspects of the data. For example, the behavior of the L_+^d mode with decreasing n in Fig. 3 is understood in terms of a decreasing contribution from the intersubband plasmon to the coupled mode. As n approaches 0 the mode converts entirely into a localized phonon with a positive-energy shift relative to the LO phonon. However, there is still some need for improvement in the model, especially in its microscopic details. Essentially, a detailed microscopic treatment is required which would treat not only the quasi-2D character of the

donor, but also the dependence of the donor's wave function, and therefore its oscillator strength, on n. This presumably would account for the difference between the calculated and measured values of L_+^d as n is reduced in Fig. 3. This will require a better microscopic understanding of the quasi-2D donor in an electron gas than is at present available.

In summary, we have measured the Raman scattering spectra of a neutral donor in a degenerate, quasi-2D electron gas in a $GaAs/Al_xGa_{1-x}As$ quantum well. New peaks in the spectrum are identified as bound plasmon–LO-phonon excitations, and are described by a simple, phenomenological theory.

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