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Dispersion of collective intersubband excitations in semiconductor superlattices

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We report the observation, by inelastic light scattering, of the dispersion of collective *inter*subband excitations of electrons confined in GaAs quantum-well superlattices. The ability to probe plasmons with large in-plane wave vectors allows one the unique possibility to study excitations in the two-dimensional limit. Intrasubband plasmons in this regime are also observed for the first time. The results are interpreted with current theories which include estimates of many-body effects.

The study of excitations in quasi-two-dimensional (2D) electron systems in surface space-charge layers and semiconductor superlattices is a subject of current interest.[1] Some of the significant experiments in such systems are associated with the observation of collective intrasubband behavior of electrons,[2-4] in which the reduced dimensionality manifests in the dispersion of the plasma frequency. In this Rapid Communication we report the first observation of the dispersion of collective *inter*subband excitations (or intersubband plasmons) in quasi-2D electron layers. The behavior of these modes, which differ from intrasubband plasmons, provides new insight into the electrodynamics of 2D electron systems. Previous experiments[3] on Si inversion layers have not revealed a wave-vector dependence of the intersubband plasmon energy. The present study is based on inelastic light-scattering measurements from GaAs quantum-well superlattices (QWS), with the ability to probe excitations with large in-plane wave vectors. Intrasubband plasma oscillations in the 2D limit are also observed for the first time in these superlattice structures.

Tselis and Quinn[5] recently presented a description of collective modes in QWS's. The model consists of parallel quasi-2D "electron-gas" planes separated by a distance d. The carriers display free-electron character for motion along the planes and are confined into subbands of energy $\hbar\Omega_i$ along z, the normal to the layers. The dispersion to order q^2, where q is the in-plane component of wave vector, including resonant screening $\alpha_{nn'}$,[6,7] and final-state $\beta_{nn'}$ (Ref. 8) interactions is given by

$$\omega_p^2 = \Omega_{0n}^2(1+\alpha_{nn}-\beta_{nn}) - [\Omega_{n0}^2\mu_{nn}S(q,k)]q + \left[\frac{\hbar\Omega_{0n}}{m}\left(1+\frac{\alpha_{nn}-\beta_{nn}}{2}\right) + v_F^2\left(\frac{3}{4}+\frac{1}{\alpha_{nn}-\beta_{nn}}\right) - \Omega_{0n}^2\gamma_{nn}\right]q^2 \quad . \tag{1}$$

In this equation v_F and $\hbar\Omega_{nn'}$ represent the Fermi velocity and energy separation between conduction subbands n and n'. The symbols μ_{nn}, γ_{nn}, and $S(q,k)$ are defined as

$$\mu_{nn} = \frac{4\pi Ne^2}{\hbar\Omega_{n0}\epsilon_s}\left|\int \Phi_n^*(z)z\Phi_0(z)\,dz\right|^2 \ ,$$

$$\gamma_{nn} = \frac{4\pi Ne^2}{\hbar\Omega_{n0}\epsilon_s}\frac{1}{6}\int dz\,dz'\,\Phi_n(z)\Phi_0(z)|z-z'|^3\Phi_n(z')\Phi_0(z') \ ,$$

$$S(q,k) = \frac{\sinh qd}{\cosh qd - \cos kd} \ .$$

$e^{i\mathbf{K}\cdot\mathbf{r}}\Phi_n(z)$ is the envelope wave function for an electron in the nth subband, N the electron areal density, ϵ_s the static dielectric constant of the layers, and k the wave-vector component normal to the planes. In polar semiconductors like GaAs collective excitations are coupled to LO phonons.[9] To include these effects, the dielectric constant is modified to a frequency-dependent function.[10]

The q independent term in Eq. (1), which is well known in inversion layers[1] and also observed in QWS's,[9] is a collective intersubband excitation with energy shifted from the subband spacing by depolarization and final-state effects. For finite q, the decrease in Coulomb energy associated with density oscillations within each layer softens the mode to order q. The electrostatic coupling between layers is accounted for by the structure factor $S(q,k)$. The term of order q^2 has three independent positive contributions, off setting in the large q limit the softening discussed above. The first two terms are related to nonvertical transitions. The largest contribution is due to the second term and is in contrast to the $\frac{3}{5}v_F^2q^2$ dispersion well known in 3D plasmas.[11] γ_{nn} is the higher-order contribution to resonant screening.

In the present inelastic light-scattering measurements, the selection of the scattering geometry shown in Fig. 1, together with unique sample design, gives access to such collective modes. q was varied by rotating the sample and the largest wave-vector transfer $q=k=(2\pi/\lambda)\eta$ occurs when scattered photons propagate parallel ($\Phi=90$, $\theta=0$) to the layers. Here η is the refractive index of GaAs at the incident wave length λ. This ability to study scattered light that propagates in a plane parallel to the interfaces was essential for observing effects related to dispersion. The GaAs-(Al_xGa_{1-x})As superlattices were grown by molecular-beam epitaxy with the normal to the layers along [001] and modulation doped[12] with Si. To further improve the efficiency of collecting photons, the superlattice was grown between two 1.6 μm (Al_xGa_{1-x})As cladding layers which act as a waveguide. For $0° < \theta < 45°$, excitations with smaller q are probed.[4] We present data from a sample which consisted of 30 periods of GaAs having well width $d = 221$ Å separated by 597 Å of $Ga_{0.77}Al_{0.23}As$ barriers. The spectra at 2 K were excited with an LD 700 dye laser operating near the fundamental optical gap. Polarization selection rules for incident and scattered beams were used for separating collective and single-particle excitations.[4]

Figure 1 illustrates, for varying q, spectra in the region of intrasubband and intersubband collective excitations. The ($\parallel$, $\parallel$) spectra with both incident and scattered polariza-

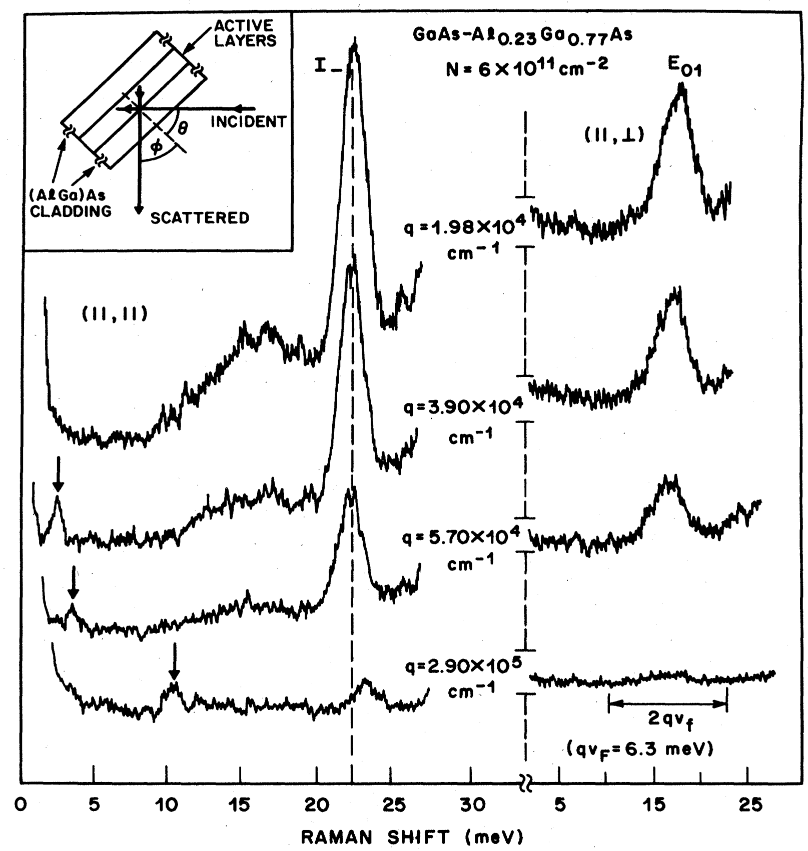

FIG. 1. Inelastic light-scattering spectra of intersubband and intrasubband (arrows) collective modes for different values of q. Coupled LO-intersubband plasmons I_- are observed with incident and scattered beam polarizations both parallel to [110], $(\parallel, \parallel)$. Single-particle excitations E_{01} appear in the crossed $(\parallel, \perp)$ configuration where scattered photons have polarizations along $[1\bar{1}0]$, orthogonal to the incident beam. The scattering geometry is shown in the inset.

tions parallel to [110] show two main features. The peak around 22 meV, labeled I_-, is identified as the lowest collective intersubband mode. The lower-energy peaks marked by arrows are attributed to intrasubband plasmons. The assignments are based upon their energy, polarization property, resonant behavior of the scattering intensity, and q dependence. The energy of the intersubband mode shows no dispersion for q up to $\approx 6\times10^4$ cm^{-1}. On changing q to the right-angle value of 2.9×10^5 cm^{-1} the sharp peak transforms to a broader band with an increase of 0.85 meV in energy. This shift in energy illustrates the first observation related to the dispersion of intersubband plasmons. The broad maximum around 15 meV in the polarized scattering has been reported earlier[13] and is related to resonant light-scattering effects. The detailed assignment of this feature is presently not well established. Single-particle excitations (SPE) across the lowest conduction subbands are also shown in Fig. 1. The difference in energy between these excitations, recorded with crossed incident and scattered photon polarizations, and the collective intersubband modes is a measure of α_{11}, the depolarization field effect. A significant broadening of the SPE is observed at large q, with corresponding decrease in intensity partly due to reabsorption. This broadening, controlled by qv_F, is insufficient to cause Landau damping of the collective excitations discussed above.

In the small-q regime $(qd < 1)$, the q-linear contribution determines the dispersion of the intersubband plasmon. For this limit, achieved in pseudo back scattering, the predicted softening in energy is small (<0.05 meV) and less than the resolution of our experiment. The observed lack of dispersion at small wave vectors is thus consistent with these theoretical estimates. Based on experimental results at $q\sim2\times10^4$ cm^{-1} $(I_-=22.1$ meV) together with a value of 15.4 meV deduced for $\hbar\Omega_{01}$ from optical emission measurements,[14] we estimate $\alpha_{11}-\beta_{11}=1.44$. The determination of the subband spacing obtained from luminescence measurements is used because those from spectra of single-particle intersubband excitations are affected by final-state interactions.[8] Contributions from the q^2 terms become appreciable for right-angle scattering $(\phi=90°)$. The small increase in the energy of I_- when q increases by over an order of magnitude is due to opposing linear and quadratic terms. Using the value for $\alpha_{11}-\beta_{11}$ deduced above, we estimate for $q=2.9\times10^5$ cm^{-1} an intersubband plasmon energy at 22.6 meV. The shift from the corresponding low q value of 22.1 meV is, thus, in good agreement with the observed increase of 0.85 meV. The energy predicted by Eq. (1) is plotted in the upper part of Fig. 2. The agreement with experiment over the range $0<q<2.9\times10^5$ cm^{-1} supports theoretical understanding of the dispersion of collective intersubband excitations in QWS's.

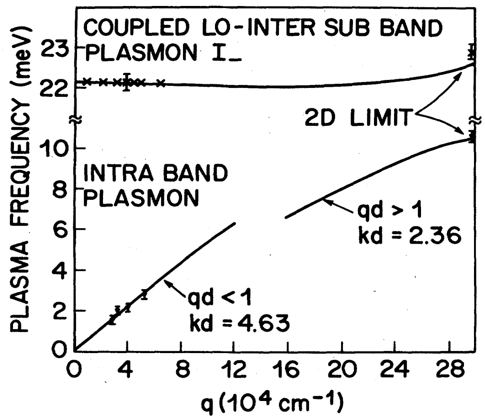

FIG. 2. (Lower): dispersion relation of collective intrasubband oscillations, including the 2D limit. The solid lines represent theoretical fits. The triangles are the experimental data. (Upper): dispersion of the collective intersubband plasmon I_-. Solid line represents I_- given by Eq. (1). Crosses are the experimental points.

The dispersion of intrasubband excitations in superlattices was observed in the small-q limit and their identification was based in part on "acoustic"-like behavior when $qd << 1$.[4] The ability to study large wave-vector modes now allows the possibility to observe intrasubband plasmons in individual layers, which for $q > 1/d$ are decoupled from neighboring electron sheets. The arrows in Fig. 1 illustrate such plasma oscillations. The peak at 10.5 meV when $q = 2.9\times 10^5$ cm^{-1} is identified as the intrasubband collective mode in the 2D limit. This mode while having the correct polarization also displays a resonance behavior similar to the lower-energy intraband plasmons. Figure 2 illustrates the calculated dispersion,[15-17]

$$\omega_p = \left[\frac{2\pi N e^2}{\epsilon_s m} S(q,k) q\right]^{1/2} ,$$

for the k values of the experiment. The linear dispersion for small q is replaced by the $\sqrt{q}$ behavior characteristic of the single layer limit $[S(qk) \to 1]$. The data are in excellent agreement with theory. This "large"-q result represents the first observation in QW superlattices of single layer behavior that has been reported in other quasi-2D systems such as electrons on the surface of liquid helium and space-charge layers.

Finally, the data also allow estimates of the final-state interaction β_{11}. Based on $\alpha_{11} - \beta_{11} \simeq 1.5$ deduced earlier and an evaluation of α_{11} from the theoretical analysis,[18] we find $\beta_{11} \simeq 0.1$. The estimate of $\alpha_{11} \sim 1.6$ was obtained within the approximations of a finite GaAs well with band bending as expressed in Ref. 18. The smallness of these many-body interactions are consistent with calculations of Ando,[8] where α_{11} is shown to strongly affect intersubband plasmons while excitonic local-field corrections for charge-density excitations cause only a weak softening.

In conclusion, we have observed the dispersion of collective intersubband excitations in QW superlattices over a wide wave-vector range. Several features of these systems are revealed for the first time in the large-q, single 2D layer regime. Our results show the importance of dispersive terms to order q^2 and provide the first verification of theoretical predictions over a large wave-vector range. Collective intrasubband plasmons have also been observed in the wave-vector regime where the superlattices display single layer behavior.

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