Discrete Plasmons in Finite Semiconductor Multilayers

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We observe discrete plasmons in layered 2D electron gases with a large, but finte, number of periods. The twofold degeneracy of plasmon modes with wave numbers in the first Brillouin zone of the infinite system is lifted by the loss of complete periodicity in the finite system. These characteristic discrete plasmon doublets are measured in inelastic-light-scattering spectra of multilayer GaAs/(AlGa)As heterostructures.

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The charge-density excitations of layered two-dimensional electron systems are the subject of current theories that consider plasma modes in semifinite[1,2] and finite[3] systems. Remarkable new effects are predicted for semiconductor superlattices. Giuliani and Quinn[1] proposed a new class of surface plasmons. They are free of Landau damping because quantization of electron motion along the superlattice axis prevents their decay into electron-hole pairs. Wu, Hawrylak, and Quinn[2] have calculated the anisotropy of charge-density excitations on the lateral surface of a semiconductor superlattice. Jain and Allen[3] have investigated the discrete plasmon spectrum of layered films. Inelastic light scattering, widely used to probe collective excitations in semiconductor superlattice,[4–9] has been suggested[1,3,10] as the experimental method to study the new charge-density excitations. This Letter reports the observation and interpretation of discrete plasmons in inelastic-light-scattering spectra of GaAs/(AlGa)As heterostructures.[11] The discrete modes are observed in multilayers with relatively large number of periods ($N \simeq 15$), which demonstrates that these effects are essential to plasma oscillations in semiconductor superlattices. In the spectra we measure characteristic plasmon doublets. They are explained by the lifting of the degeneracy of pairs of modes with wave numbers q_β and $2\pi/d - q_\beta$ in the first Brillouin zone of the superlattice with period d. In the infinite superlattice these modes are degenerate because each 2D electron gas is at a plane of mirror symmetry. The lifting of the degeneracy in finite multilayers is due to the loss of these symmetries.

A superlattice of 2D electron gases each with an areal density n embedded in a material with dielectric constant ϵ_S has a plasma frequency[12–16]

$$\omega_p^2(q,q_\beta) = \omega_0^2(q) S(q,q_\beta), \tag{1}$$

where q is the component of wave vector in the plane parallel to the layers. With $N \to \infty$,

$$q_\beta = \beta \frac{2\pi}{Nd}, \quad \beta = 1, \ldots, N, \tag{2}$$

is the perpendicular wave number and d is the superlattice period. $\omega_0(q)$ is the 2D plasma frequency,[17]

$$\omega_0^2(q) = 2\pi n e^2 q / m^* \epsilon_S + 0.75 q^2 v_F^2,$$

where m^* is the electron effective mass and v_F is the Fermi velocity.

$$S(q,q_\beta) = \sinh qd / (\cosh qd - \cos q_\beta d)$$

is a structure factor for the superlattice.

For finite N the discrete plasmon modes are obtained by the coupling among oscillations with frequencies $\omega_p(q,q_\beta)$.[3,14] When the layers are embedded in an infinite dielectric the ϵ-ϵ-ϵ geometry of Ref. 3 is applicable and there are no surface modes. The equations for the plasmon frequencies are derived from Eq. (7) of Ref. 3 by setting $a_1 = a_3$. We obtain two *separate* sets of modes that are the solutions of[18]

$$\frac{1}{\omega_0^2(q)[\cosh qd \pm 1]} = J \sum_{\beta=1}^{N} \frac{b^{\pm}(q,q_\beta)}{\omega_\pm^2 - \omega_p^2(q,q_\beta)}, \tag{3}$$

where

$$b^{\pm}(q,q_\beta) = \frac{\pm [1 \mp \cos q_\beta d]}{2N[\cosh qd - \cos q_\beta d]^2} \tag{4}$$

and $J = 1 - \exp(-Nqd)$. The existence of two separate sets of modes is due to the lifting of the twofold degeneracy of the superlattice plasmons in the finite system.

Equations (3) and (4) yield the frequencies of the plasmon pairs for all values of N. For values of q_β such that the spacings between $\omega_p(q,q_\beta)$ for consecutive values of β is larger than $2\omega_0^2(q) \times [\cosh qd \pm 1] b^{\pm}(q,q_\beta)$, the mode frequencies can be calculated within the first-order approximation

$$\frac{1}{\omega_0^2(q)[\cosh qd \pm 1]} \simeq J \frac{2b^{\pm}(q,q_\beta)}{\omega_\pm^2 - \omega_p^2(q,q_\beta)}. \tag{5}$$

In this limit the major effect of the finite number of layers is the coupling of the degenerate pair with wave vectors q_β and $q_{N-\beta} = 2\pi/d - q_\beta$. It results in pairs of modes that are symmetric and antisymmetric combinations with frequencies

$$\omega_\pm^2(q,q_\beta) = \omega_p^2(q,q_\beta) + \omega_0^2(q) F_\pm(q,q_\beta), \tag{6}$$

where

$$F_{\pm}(q,q_\beta) = J\frac{[1-\cosh qd\,\cos_\beta d] \pm [\cosh qd - \cos q_\beta d]}{N[\cosh qd - \cos q_\beta d]^2}. \quad (7)$$

This approximation gives exactly the same number of different modes as in the more exact expressions of the ϵ-ϵ-ϵ geometry of Ref. 3 and of Eqs. (3) and (4).

Inelastic–light-scattering experiments have verified the bulk plasmon dispersion of Eq. (1) in multiple GaAs/(AlGa)As heterostructures with $15 \leq N \leq 20$.[5,9] In the results reported here we discovered characteristic plasmon doublets described by Eqs. (6) and (7) in GaAs/(AlGa)As multilayers with $N = 15$. The multiple plasmon peaks are best resolved in spectra measured with wave vectors near the "long wavelength" limit, in which the $q_\beta d$ values are closer to 2π.

We studied several modulation-doped[19] multiple GaAs/(Al_xGa_{1-x})As quantum well heterostructures. We present the data from an n-type sample in which the GaAs wells have a thickness $d_1 = 258$ Å and the ($Al_{0.24}Ga_{0.76}$)As barriers have $d_2 = 520$ Å. Fifteen periods of $d = d_1 + d_2 = 778$ Å were grown on GaAs(001) substrates by molecular-beam epitaxy. The free-electron density is 4×10^{11} cm^{-2} and their mobility is $\mu = 9\times10^4$ cm^2/V·sec. We have determined a lowest subband spacing of 13 meV. The concentration of carriers in the first excited subband is less than 0.2×10^{11} cm^{-2}. For these parameters the electron density peaks at the center of the quantum well.[20]

The spectra were excited with tunable dye lasers operating in the cw mode in the wavelength range 6300 Å $< \lambda_L <$ 7800 Å. We report the results obtained with wavelengths of 6760 and 7630 Å. In both cases the photon energies, 1.8342 and 1.6251 eV, are in resonance with optical transitions of the GaAs quantum wells. Low-temperature light-scattering spectra were obtained in the nearly backscattering geometry of Olego *et al.*[5] and also in conventional backscattering. The in-plane component of the scattering wave vector was tuned in the range $3\times10^4 < k < 18\times10^4$ cm^{-1} by changes in the angle between the normal to the layers and the incident laser beam. The perpendicular component k_z was changed by variation of the incident laser wavelength.

Figure 1 shows spectra excited with $\lambda_L = 6760$ Å. The top spectrum was obtained in conventional backscattering and the two lower ones in nearly exact backscattering. We are interested in the structures labeled S (for strong) and W (for weak). The positions of these peaks have a strong dependence on the in-plane scattering wave vector k. This is unambiguous evidence that they arise from plasma oscillations of the free electrons in the multilayers. Weaker structure can be seen at lower and higher energies. The spectra show also a broad scattering (between 7 and 15 meV) that has no k dependence. The multiple structures in Fig. 1 indicate plasma modes with well-resolved discrete character. The stronger peaks are doublets, which suggests that the multiplicity is due to the finite-system effects described by Eqs. (6) and (7). In fact, we show below that the measured plasmon frequencies are interpreted by means of Eqs. (6) and (7) with the q_β value that is closest to k_z.

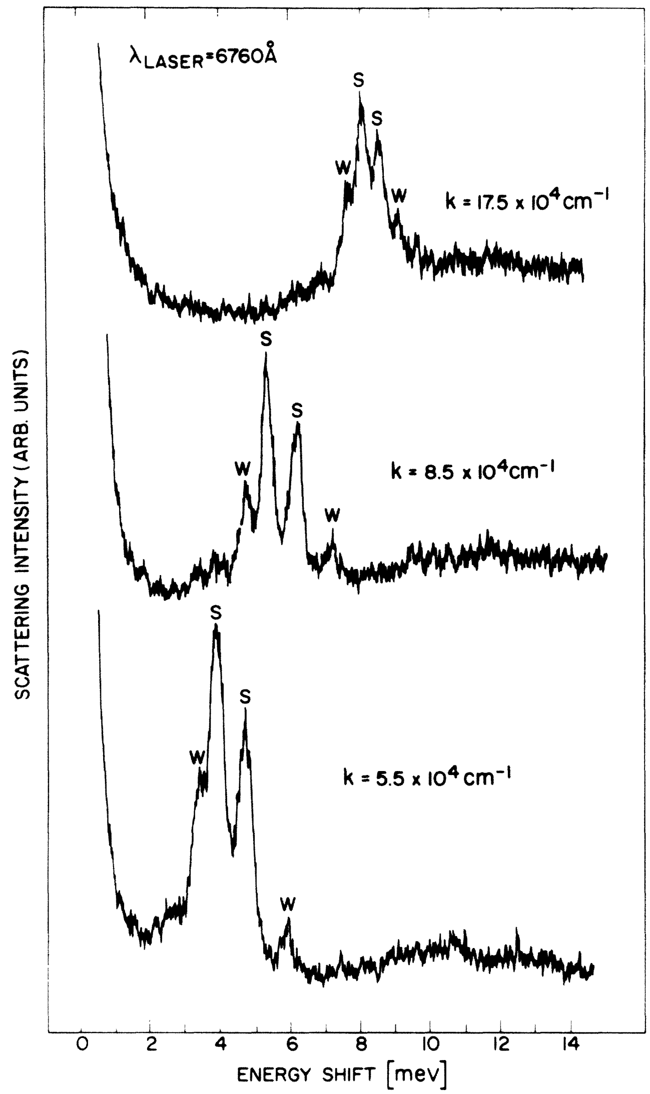

FIG. 1. Inelastic–light-scattering spectra of discrete plasmons taken at different values of the in-plane scattering wave vector k. The spectra were excited with a laser wavelength of $\lambda_L = 6760$ Å.

The spectra in Fig. 1 are strikingly different from the single broad band previously reported.[5] The spectral differences arise from the larger value of k_z at

$\lambda_L = 6760$ Å. For an estimated[21] refractive index of $\eta = 3.67 \pm 0.02$ we obtain $2k_L = 4\pi\eta/\lambda_L = 6.82\times10^5$ cm^{-1}. It yields $k_z = 2k_L[1-\frac{1}{2}(k/2k_L)^2]$ for conventional backscattering and

$$k_z = k_0[1-(1/2\eta)^2] \simeq 6.7\times10^5\ \text{cm}^{-1}$$

in nearly exact backscattering. The value $k_z d \simeq 5.25$ being closer to the "long wavelength" limit ($k_z d = 2\pi$) than those of Olego *et al.*[5] allows coupling to discrete plasmon doublets of wider separation. This interpretation is supported by the spectra of Fig. 2 obtained with $\lambda_L = 7630$ Å. In this case the lower value of $k_z d \simeq 4.55$ allows scattering by plasma modes with less sensitivity to finite system effects. In Fig. 2 the k-dependent structure of the multilayer plasmon is resolved only for the larger values of k. The stronger features appear as a doublet, further evidence of finite system splittings predicted by Eqs. (5)–(7).

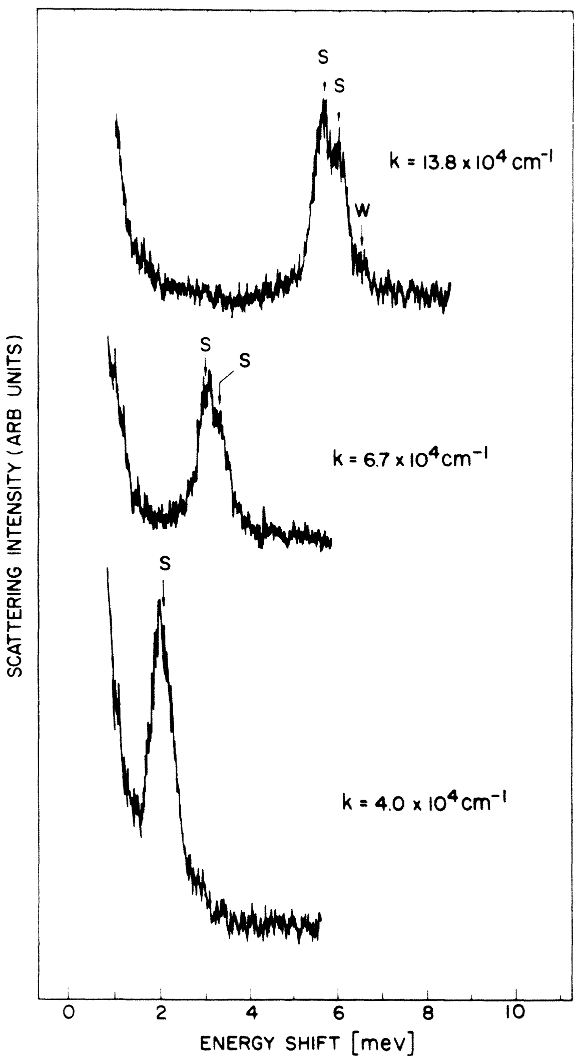

FIG. 2. Typical light-scattering spectra of discrete plasmons obtained with $\lambda_L = 7630$ Å.

Figure 3 shows the dispersions of the major plasmon peaks, strong and weak, measured in the light-scattering spectra. For $\lambda_L = 6760$ Å, with the larger value of k_z the plasmon dispersion displays a striking departure from the nearly linear, acousticlike dispersion at the smaller value of k_z. In these results the finite-N effect is represented by about one-half the spacings between the two strong peaks. On the other hand, the separations between modes of consecutive values of β are given by the spacings between the high-energy weak and low-energy strong peaks (or, equivalently, by the spacings between the low-energy weak and high-energy strong peaks). In the quantitative interpretation of the measured dispersions we assume the ϵ-ϵ-ϵ geometry of Jain and Allen.[3] This geometry is used because the top layer is a 520-Å-thick $Al_{0.24}Ga_{0.76}As$ barrier with a dielectric constant ϵ_S nearly identical to that of the GaAs quantum wells. We see in Figs. 1 and 3 that the separations between modes of consecutive values of β are considerably larger than the finite-N effects, and we conclude that Eqs. (6) and (7) represent a reasonable first-order approximation to interpret our experiments.

The plasmon dispersions calculated with Eqs. (6) and (7) are very sensitive to the number of layers N. To fit the measured dispersions we assume in-plane wave-vector conservation ($k = q$) and adjust the value of N. Best agreement is obtained with $N = 14$, one fewer than the number of periods in the sample.[22] The greatest sensitivity to the value of N occurs for $0.6 \leqslant q \leqslant 1.2\times10^5$ cm^{-1}. In this range, the frequen-

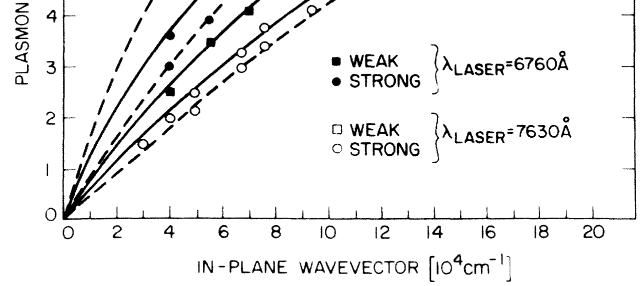

FIG. 3. The points are the peak positions in light-scattering spectra of discrete plasmons plotted as a function of the in-plane scattering wave vectors. The lines are the calculated discrete plasmon dispersions.

cies calculated with $N=15$ are 0.2 meV higher than those with $N=14$ and those with $N=13$ are 0.2 meV lower. This result indicates that the top GaAs layer is depleted of free electrons. It is not surprising since GaAs samples have depletion surface–space-charge layers.[23] In Fig. 3 the full and dashed lines are the high (antisymmetric) and low (symmetric) energy components of the doublets predicted by Eqs. (6) and (7) for $N=14$. There is good agreement with measured frequencies. The small differences at the highest values of in-plane wave vector could indicate corrections in the 2D plasma frequency due to finite width of the electron envelope function,[24] and coupling to intersubband excitations.[25]

The agreement between measured and calculated dispersions implies that for $N=14$ the major effect of the finite number of layers is the lifting of the degeneracy of the infinite system. In addition to the strong doublets with $q_\beta \approx k_z$, the spectra also show weaker peaks of modes with adjacent values of perpendicular wave vector. These could be explained by the lack of conservation of the perpendicular component of wave vector in the finite multilayer systems. For photon absorption lengths much larger than d, the intensity of light scattered by each discrete mode is[16]

$$I_\beta \sim \frac{1}{1-\cos[(q_\beta - k_z)d]}. \tag{8}$$

Equation (8) is in reasonable agreement with the relative intensities of weak and strong features. A complete theory of light-scattering intensities should consider the interference between the β and $N-\beta$ components of the charge-density fluctuations of each mode, and terms beyond the first-order approximation represented by Eqs. (6) and (7) that would add a small admixture with other q_β values. The interference could explain the small ($\sim 20\%$) differences in intensities between the low-energy (symmetric) and high-energy (antisymmetric) plasmon peaks. The finite width of the modes also has to be taken into account. In the best resolved spectra we measure a width of 0.3 meV, about 50% larger than the width obtained from the electron mobility ($\gamma = e/m^*\mu$).

The surface depletion layer has important consequences for the existence of Giuliani-Quinn[1] surface plasmons. Well-defined surface modes exist when $[(\epsilon_S - 1)/(\epsilon_S + 1)]\exp(-2qd') \simeq 1$,[1,16] where d' is thickness of the surface depletion layer. In our multiple heterostructures we estimate $d' \approx 1000$ Å, and well-defined surface plasmons may not exist. Inelastic–light-scattering experiments in search of surface plasmons in samples of special design are in progress.

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