Many-body coupling between quasiparticle and collective excitations in semiconductor quantum wells

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We predict that, due to a many-body coupling between collective and single-particle excitations, there is an experimentally observable resonant line splitting in the intersubband collective charge-density excitation spectra in wide semiconductor quantum wells at low electron densities. This many-body coupling causes even the long-wavelength intersubband plasmon mode to be Landau damped at a critical density. Above the critical density, the intersubband plasmon mode lies *lower* in energy than the single-particle excitation.

Elementary excitations in an electron gas are traditionally decomposed into two distinct categories:[1-3] single-particle electron-hole excitations and collective plasmon modes. The single-particle excitations correspond to the excitation of an electron above the Fermi surface leaving a hole below, whereas the plasmon is a collective charge-density excitation of the whole system arising from the long-range nature of Coulomb interaction. In the interacting Fermi system, this basic picture still holds due to the validity of the Landau Fermi-liquid theory[1-3] in which single-particle excitations are interpreted as quasiparticles. At low excitation energies, these quasiparticles are long lived and the interacting Fermi liquid consists of weakly interacting quasiparticle excitations and a collective plasmon mode which, at long wavelengths, has the same energy as the bare plasmon due to the f-sum rule.[1-3] At large wave vectors, the plasmon mode enters the electron-hole continuum of single-particle excitations and becomes Landau damped due to its decay into electron-hole pairs. Thus the distinction between collective and quasiparticle excitations is lost at large wave vectors. This paradigm of the elementary excitation spectra of a quantum solid-state plasma consisting (at least, for long wavelengths) of low-energy quasiparticles and (a high-energy) collective plasmon mode has long served as the basic model for normal metals in three dimensions. More recently, this paradigm has been used very successfully[4] in describing the properties of two-dimensional electron-gas (2DEG) systems confined in semiconductor inversion layers, quantum wells, and heterostructures. It is not an exaggeration to state that this paradigm of a normal Fermi liquid (in two- and three-dimensional systems) being composed of quasiparticles and plasmons, which are distinct from each other at long wavelengths, is one of the most well-established notions in condensed-matter physics. During the last four decades, this paradigm has been the basis for interpreting a wide range of experimental techniques which probe electronic properties of solids, including inelastic light and electron-scattering spectroscopies, optical and photoemission spectroscopies, etc. In this paper, we make quantitative theoretical predictions about an experimentally realizable low-density situation where this distinction fails even at long wavelengths due to a many-body coupling between single-particle and collective excitations. In particular, we show that, under suitable conditions, there are situations in a 2DEG where single-particle excitations and plasmons are *not* distinct excitations, but are strongly coupled together by a many-body effect which should lead to an observable resonant line-splitting phenomenon in inelastic light-scattering and far-infrared absorption spectroscopic experiments capable of probing the charge-density excitations in semiconductor quantum wells. The remarkable feature of our prediction is not just that there is a many-body coupling between plasmons and quasiparticles, but that it can exist even at long wavelengths under suitable experimental conditions. To the best of our knowledge, the possibility of a *long-wavelength* many-body coupling between single-particle and collective excitations in an electron gas has never before been discussed in the literature.

To qualitatively describe this resonant coupling effect, we start by considering a two-dimensional (2D) structure where the electrons are free to move along the x-y plane, but their motion along the z direction is quantum mechanically confined by potential barriers. This leads[4] to the formation of quantum subbands associated with z motion, and the single-particle electron wave function can be denoted by $(i,\mathbf{k})$, where $\mathbf{k}$ is the conserved 2D wave vector associated with the free motion in the x-y plane and i is the subband index associated with the quantum subband for z motion. In this subband-quantized 2DEG, there are two different kinds[4,5] of elementary excitations associated with intrasubband motion (where the subband index i is conserved) and intersubband (where i changes) transitions. Both intrasubband and intersubband elementary excitations have been extensively experimentally studied[6] in 2DEG using far infrared and light-scattering spectroscopies. The subject matter of this paper are the intersubband elementary excitations in low-density, wide quantum wells where a resonant many-body mode-coupling phenomenon between intersubband single-particle excitations (SPE) and collective (plasmon) charge-density excitations (CDE) take place.

In a confined 2D structure, where the confinement potential along the z direction produces quantum subbands for the z motion and the motion along the x-y plane is free, the single-particle energy levels are given by (using atomic units $\hbar=2m=1$, where m is the electron effective mass) $E_i(k)=E_i+k^2$, where i is the subband index and $\mathbf{k}$

is the free 2D wave vector, and E_i the ith subband bottom energy. (Throughout, we assume isotropic, parabolic bands with the z and the x-y motions totally decoupled.) Transitions from a subband state $(i,\mathbf{k})$ to another subband state $(j,\mathbf{k}+\mathbf{q})$ is an intersubband transition provided that $i \neq j$. The SPE intersubband transition is given by the single-particle energy difference $E_{ji}(\mathbf{k},\mathbf{q}) \equiv E_j(\mathbf{k}+\mathbf{q}) - E_i(\mathbf{k}) = E_{ji} + q^2 + 2\mathbf{k}\cdot\mathbf{q}$, where $E_{ji} \equiv E_j - E_i$ is the subband separation. In addition to the SPE intersubband transition, one may excite intersubband collective CDE's, where the transition between the two subbands is dynamically screened by all the other electrons in the system and is therefore associated with a macroscopic polarization of the 2D layer along the z direction, and is consequently plasma shifted above the single-particle intersubband energy by a depolarization shift.[6-11] There is, in addition, an excitonlike, many-body vertex correction[9-11] to the collective intersubband mode associated with the Coulomb interaction between the electron in the upper subband and the hole left behind in the lower subband which tends to lower the intersubband transition energy below the single-particle intersubband energy. The relative resonance energies of SPE and CDE depend on the details (electron density, confinement potential, etc.) of the system, but, in general, CDE tends to be higher in energy than the SPE, because, in the situations studied so far, vertex correction is usually smaller than the depolarization shift.

In this paper, we consider intersubband transitions in quantum wells as a function of the 2D wave vector (q), emphasizing the low electron-density regime where exchange-correlation-induced many-body effects are relatively more important quantitatively. We include exchange-correlation effects in our theory through the usual[12] local-density approximation[13] (LDA)—we have, however, tried some nonlocal forms[14] for the exchange-correlation potential in our calculation, finding very little difference (less than a few percent) with the LDA results presented here. In our theory we include exchange-correlation corrections self-consistently, both in the calculation of the subband energy levels and the wave functions as well as in the linear-response calculation. We use a coupled self-consistent linear-response–local-density-functional formalism to calculate the intersubband resonance to the external perturbation. We start from a fully self-consistent local-density-functional calculation of the subband energies, where the exchange-correlation corrections to the single-particle energy levels are included in the LDA (Ref. 13) which has earlier been successfully employed[12] for 2D GaAs structures. Then we carry out a finite wave-vector linear-response calculation using these LDA energy levels and wave functions where the first-order change in the Hartree (ΔV_H) and the exchange-correlation (ΔV_{xc}) potentials due to the modification of the electron-density distribution by the external perturbation is included in the self-consistent potential. Using time-dependent perturbation theory, we calculate the dynamical conductivity or, equivalently, the intersubband dielectric function of the system from which one can obtain the far-infrared absorption or the Raman-scattering spectrum and the intersubband transition energies. In this theory, ΔV_H and ΔV_{xc} represent the depolarization shift and the excitonic correction, respectively, and, by keeping both of them in the calculation one obtains the CDE energies, whereas by setting $\Delta V_H = \Delta V_{xc} = 0$, one obtains the SPE energies. Note that the self-consistent theory employed here is diagrammatically equivalent to keeping the ladder diagrams for vertex correction (excitonic shift) and the ring diagrams for resonance screening (depolarization shift) with the exchange-correlation self-energy corrections to individual energy levels being calculated in the LDA. Since the calculation is done self-consistently, current conservation is manifestly obeyed and, therefore, the approximation scheme is conserving. Our calculational scheme is thus roughly equivalent to the time-dependent Hartree-Fock theory for linear response (i.e., the ladder-bubble diagrams), except that the exchange-correlation effect is treated in the LDA. This approximation is occasionally referred to as the time-dependent local-density approximation (TDLDA).

Before presenting our results, it is worthwhile to discuss the connection between the current work and earlier theoretical work on the subject. Our theory is a direct finite wave-vector generalization of Ando's long-wavelength ($q = 0$) calculation[9] of intersubband optical absorption in Si space-charge layers. If one leaves out the exchange-correlation potential completely from the theory (i.e., both from the single-particle energy-level calculations of the density-functional theory and from the linear-response calculation), one gets the extensively used random-phase approximation (RPA) or the time-dependent Hartree approximation, which in this particular context was first worked out[7] by Allen, Tsui, and Vinter in the long-wavelength limit.

There have been a number of recent calculation[15-17] of intersubband response in 2D GaAs microstructures using approximation schemes which are equivalent to ours. Eliasson, Hawrylak, and Quinn[15] carried out a calculation of inelastic light-scattering spectra by collective charge-density excitations in *multilayer superlattices* using a LDA-linear-response theory which is equivalent to our quantum-well TDLDA calculation. Gammon and co-workers[16,17] carried out a LDA self-consistent-field calculation, which is equivalent to our TDLDA theory, for spin-density excitations[16,17] and for CDE's (Ref. 17) in GaAs square wells, comparing the numerical results with their experimental inelastic light-scattering spectra (and obtaining very good agreement). There have also been two[18,19] recent attempts at calculating the collective-mode spectra in 2D quantum wells by including nonlocal exchange in the theory within a Hartree-Fock approximation. These calculations,[18,19] which are equivalent to the time-dependent Hartree-Fock theory, find[17] that the TDLDA calculations are in fact quantitatively very accurate (better than 5%) except at the highest electron densities. Our interest primarily being at low electron densities, TDLDA should be a very good approximation for our results. We emphasize that none of these[7-11,15-19] references, spanning over a range of more than 15 years and two classes of systems (i.e., Si inversion layers and GaAs heterojunctions), alluded to the possibility of obtaining a collective intersubband CDE which is actually

below the intersubband SPE and of the many-body resonant-coupling line-splitting phenomenon predicted in this paper.[4–11,15–20]

In the rest of this paper, we present and discuss our numerical results for the lowest intersubband mode in the system. (We compare TDLDA and RPA results to emphasize the role of exchange-correlation effects in the former.) All our calculations are at $T=0$ for realistic GaAs quantum-well systems confined by $Al_xGa_{1-x}As$ barriers within the effective-mass approximation and in the electric quantum limit (i.e., under the conditions where only the lowest quantum subband is occupied) within a two-subband approximation. We have carried out calculations for both square and parabolic quantum wells for a wide range of electron densities (N_S) and confinement parameters (e.g., the width of the well, etc.). All our energies are in GaAs atomic units (Ry) with 1 Ry=5.8 meV and the Bohr radius $a_B=98.7$ Å.

In Fig. 1, we first show our TDLDA-calculated dynamical structure factor for far-infrared absorption or Raman-scattering spectra which directly measures the intensity of the intersubband CDE mode. The results are for a parabolic well corresponding to Ref. 14 with electron densities $N_S=0.15\times10^{11}$ cm^{-2} [Figs. 1(a) and 1(b)] and 0.5×10^{11} cm^{-2} [Figs. 1(c) and 1(d)]. The resonant line splitting due to many-body coupling between CDE and SPE modes is clearly visible in Figs. 1(a) ($q=2\times10^4$ cm^{-1}) and 1(b) ($q=5\times10^4$ cm^{-1}), in spite of the fact that we have used a rather large impurity-induced level broadening corresponding to a low mobility of 10^5 cm^2/V s in our calculations. In higher-mobility samples, the line splitting is more prominent. In the high-density sample, the line splitting does not show up [Figs. 1(c) and 1(d)] and, in fact, for a large wave vector $q=3\times10^5$ cm^{-1} [Fig. 1(d)], the CDE mode is highly damped because it is inside the SPE continua. The result shown in Fig. 1(c) is the usual expected intersubband plasmon dynamical structure factor with a well-defined (and level-broadened) plasmon peak. The results shown in Figs. 1(a) and 1(b), which show up only in the finite wave-vector TDLDA calculation (and *not* in the RPA) for low electron densities (around $1-2\times10^{10}$ cm^{-2}), are the principal predictions of our work.

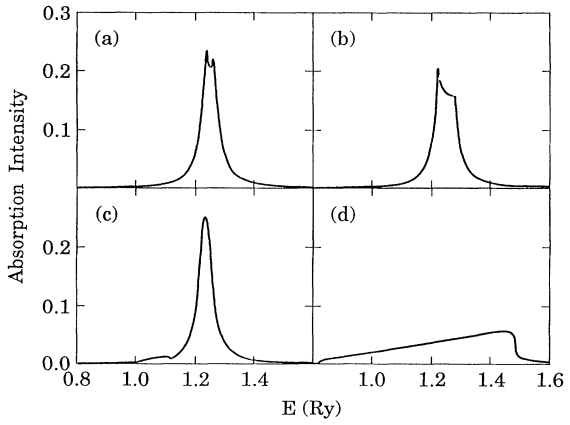

FIG. 1. The LDA-calculated far-infrared absorption intensity (in arb. units) in the parabolic well for (a) $N_S=0.15\times10^{11}$ cm^{-2} and $q=2\times10^4$ cm^{-1}; (b) $N_S=0.15\times10^{11}$ cm^{-2} and $q=5\times10^4$ cm^{-1}; (c) $N_S=0.5\times10^{11}$ cm^{-2} and $q=5\times10^4$ cm^{-1}; and (d) $N_S=0.5\times10^{11}$ cm^{-2} and $q=3\times10^5$ cm^{-1}. (c) is the usual undamped CDE resonance, whereas (d) is the highly damped situation. In (a) and (b), the many-body line splitting is clearly visible. The sample has a rather low mobility of 10^5 cm^2/V s.

To understand the many-body line-splitting phenomenon, in Fig. 2 we show our calculated intersubband-mode energy dispersion as a function of the 2D wave vector q for a parabolic well with $N_S=1.1\times10^{11}$ cm^{-2} (a), 0.2×10^{11} cm^{-2} (b), and 0.1×10^{11} cm^{-2} (c). As insets of Fig. 1, we show our cal-

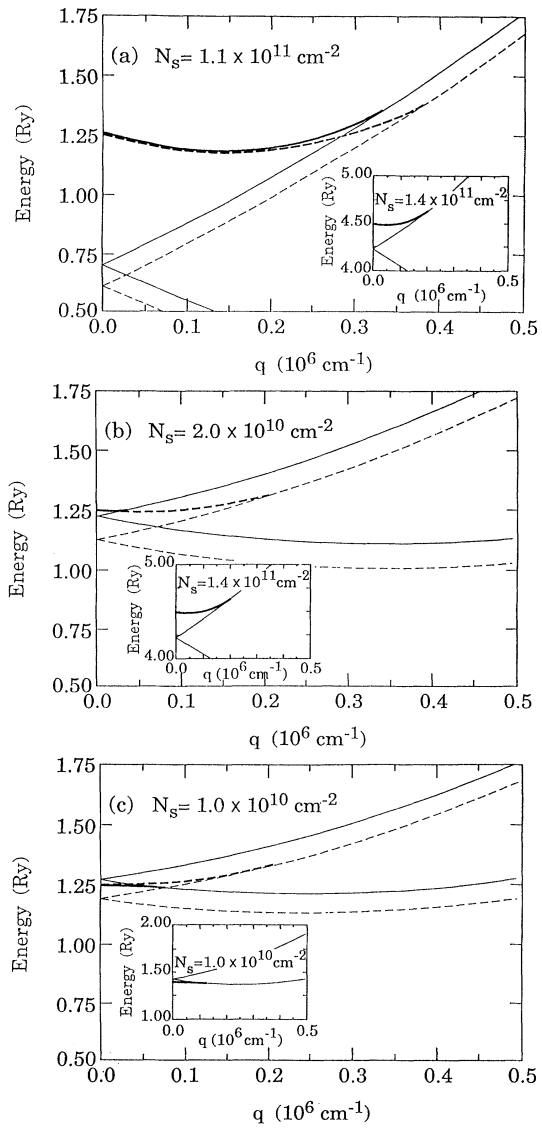

FIG. 2. The lowest calculated intersubband CDE (bold lines) and SPE (thin lines) transition energies in a GaAs-$Al_xGa_{1-x}As$ parabolic well of 1600-Å width as a function of the 2D wave vector for three different electron densities as shown. The solid lines are self-consistent LDA results and the dashed lines are RPA results. The insets show the LDA results for the intersubband transition energies (CDE and SPE) for GaAs-$Al_xGa_{1-x}As$ square wells with 400- [(a) and (c)] and 200-Å (b) widths.

culated square-well results with the width $W=400$ Å, $N_S=1.4\times10^{11}$ cm^{-2} (a) and 0.1×10^{11} cm^{-2} (c), and $W=200$ Å, $N_S=1.4\times10^{11}$ cm^{-2} (b). All our square-well results are based on a finite well with a barrier $V_0=75$ Ry. For the parabolic wells, we choose the well width to be parabolic up to 1600 Å with the confining potential a truncated parabola corresponding to the experimental samples used in Ref. 20. In Fig. 2, we show (for the parabolic wells) both the TDLDA (solid) and the RPA (dashed) results and both the CDE (bold) and the SPE (light) intersubband dispersion. For square wells, we show only the full TDLDA results. An interesting feature of our results is the negative dispersion exhibited by the intersubband CDE in the low-q region (persisting up to about $q\approx2\times10^5$ cm^{-1} at higher N_S) with a shallow minimum in the CDE dispersion around a characteristic wave vector (q_c) which depends on the electron density (and is around $1-3\times10^5$ cm^{-1}). We find this intersubband dispersion minima both in the RPA and in the TDLDA, and for both parabolic and square wells. The minimum is stronger for wider wells and at higher electron densities. The size of the minimum and its location q_c are such that they should be observable in Raman-scattering and far-infrared spectroscopic experiments.

In addition to the dispersion minimum, the results in Fig. 2 show two interesting nonlocal *many-body* features. One is the interplay between the intersubband CDE collective mode and the SPE Landau continua. The CDE mode is damped beyond the SPE Landau continua and, therefore, the sharp collective mode disappears at the critical wave vector where it intersects the SPE line. This critical wave vector associated with the vanishing of the intersubband CDE collective mode is much smaller for the many-body TDLDA theory than for the RPA. Thus experimentally one expects the intersubband collective mode to be overdamped at wave vectors substantially lower than the RPA prediction. The effect is particularly pronounced at low values of N_S, where the many-body CDE vanishes at very small wave vectors, but the RPA theory predicts persistence of the CDE up to fairly large values of wave vectors.

The second interesting many-body feature of our results is that, at low enough electron densities, the CDE mode can actually lie *below* the single-particle continua. This is a pure many-body effect arising from the excitonic vertex correction becoming larger than the depolarization shift at low electron densities. Obviously, this effect does not exist within the RPA, which neglects vertex corrections completely. Thus an experimental observation of the intersubband CDE mode being below the intersubband SPE will be clear evidence for a many-body phenomena. Our results show this to be experimentally feasible both in the parabolic and square quantum wells at $N_S\sim1-2\times10^{10}$ cm^{-2}. This is the only situation known to us where a collective mode actually lies *below* a single-particle continuum.

One curious consequence of the above-mentioned many-body interplay between the CDE and the SPE modes is the existence of a critical electron density N_C (for a given sample configuration) at which the CDE and the SPE are degenerate at $q=0$, leading to the (almost) vanishing of even the long-wavelength intersubband plasmon mode in quantum wells. This happens around an electron density of 0.15×10^{11} cm^{-2} ($\approx N_C$) for most of the parabolic wells we have investigated. Direct calculation (Fig. 1) of the infrared-absorption and the Raman-scattering spectra shows the remarkable long-wavelength resonant line-splitting phenomenon around N_C due to the many-body coupling between the collective CDE and the single-particle SPE modes. Above the critical density, the CDE plasmon has higher energy than the SPE continua, and below the critical density the CDE modes lie lower in energy than the SPE. Note that, as N_S is lowered, many-body repulsion between the SPE Landau continua and the CDE plasmon mode pushes the wave vector q_c (where the intersubband CDE has a minimum) to lower values, and, precisely at $N_S=N_C$, the minimum disappears because $q_c=0$. For $N_S<N_C$, there is no longer a minimum in the intersubband CDE dispersion, and, in fact, the long-wavelength plasmon dispersion is non-negative for $N_S<N_C$. Thus the curvature of the intersubband CDE plasmon dispersion at long wavelengths changes as one goes through $N_S=N_C$ and pushes the plasmon mode below the Landau continuum.

Let us emphasize that these many-body features, namely the vanishing of the intersubband CDE plasmon at $N_S=N_C$, the resonant line splitting, the plasmon energy being lower than the single-particle energy for $N_S<N_C$, and the change in the slope of the long-wavelength CDE dispersion around $N_S\sim N_C$ are all purely exchange-correlation effects which do not exist within the RPA. (We point out that our TDLDA response calculation should be well valid[17] at the electron densities considered in this paper.) We believe that these many-body effects should be observable[6,16,17] in a Raman-scattering or far-infrared absorption experiment[6,20] where our predicted line splitting of Fig. 1 should show up strikingly. Varying the electron density in the well with a remote gate, it should be possible to see the vanishing of the long-wavelength plasmon mode at $N_S=N_C$ and also the pushing of the collective mode below the SPE continua.

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