Interplay between disorder and intersubband collective excitations in the two-dimensional electron gas

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Intersubband absorption in modulation-doped quantum wells is usually appropriately described as a collective excitation of the confined two-dimensional electron gas. At sufficiently low electron density and low temperatures, however, the in-plane disorder potential is able to damp the collective modes by mixing the intersubband charge-density excitation with single-particle localized modes. Here we show experimental evidence of this transition. The results are analyzed within the framework of the density functional theory and highlight the impact of the interplay between disorder and the collective response of the two-dimensional electron gas in semiconductor heterostructures.

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Intersubband (IS) excitations of two-dimensional electron gases (2DEGs) confined in semiconductor heterostructures are among the most important probes of electron-electron interactions and represent an active area of research. Further motivation for the experimental and theoretical study of IS transitions stems from the growing number of opto-electronic devices whose operation is based on these transitions. In the last decade several authors have demonstrated that –particularly in the long-wavelength limit– IS charge-density modes (the ones probed in optical absorption experiments) are significantly shifted from the single-particle transition energies by both direct (depolarization shift) and exchange-correlation (excitonic coupling) terms of the Coulomb interaction. The influence of these dynamic contributions to the elementary excitation spectrum of 2DEGs has been studied experimentally and theoretically modeled. Only in recent times, however, attention has been given to the interplay between disorder and many-body contributions. This interplay raises several fundamental questions on the role of dephasing and scattering in determining the collective response of electrons and has direct implications on the optimization of IS-based devices.

Metzner et al. and Ullrich et al. addressed the influence on the IS linewidth of the various scattering mechanisms and developed microscopic theories able to treat many-body effects and disorder on equal footing. One of the main results of these models is the precise description of how the in-plane disorder destroys the coherence of the collective excitations and modifies the IS linewidth and peak energy. In particular, Metzner et al. emphasized that a transition from a regime where the intersubband absorption is rather broad and originates from randomly distributed localized single-particle excitations to a regime dominated by a sharp and blue-shifted collective mode of the 2DEG can occur by varying the electron density. This effect was explained as a mutual phase adaptation of the localized oscillators interacting via long-range Coulomb forces. Recently Yakimov et al. reported experimental evidence of such a transition in the case of holes in a self-aggregated Si-Ge quantum dot matrix.

In this article we report a study of mid-infrared IS excitations in 2DEGs confined in modulation-doped narrow GaAs/Al0.3Ga0.7As quantum wells (QWs) under the application of a perpendicular electric field. The latter allows us to vary the carrier sheet density in the range 109–1012 cm−2 and explore regions where the relative role of disorder and many-body effects are expected to be markedly different without reaching the extreme localization limit studied by Yakimov et al. in Ref. [4]. We present quantitative analysis of IS transitions based on a model that includes both static and dynamic many-body effects. Our measurements reveal a substantial softening of the IS excitation energy accompanied by an abrupt increase of the intersubband absorption linewidth. This behavior is observed at low temperatures and at electron densities ∼1011 cm−2. We shall argue that at these densities a transition occurs from a regime dominated by the collective response of free electrons to another driven by the influence of localized single-particle intersubband modes on this response. This is analogous to the Landau damping mechanism occurring at finite in-plane wavevector in a disorder-free 2DEG.

The two samples used for this study are modulation-doped single GaAs/Al0.3Ga0.7As QWs (8.7-nm- and 7.5-nm-thick, respectively). Doping in the Al0.3Ga0.7As layers was offset by about 25 nm from the wells, and the growth was performed by molecular beam epitaxy. At equilibrium samples have free-electron concentrations close to 1012 cm−2 and low-temperature mobility above 105 cm2/Vs. Well thickness was chosen in order to avoid population of the second subband and to maximize the impact of fluctuations due to interface roughness, while preserving high values of electron mobility. Measured low-temperature IS transitions peak at 112.5 meV (8.7-nm-thick QW) and 130.5 meV (7.5-nm-thick QW). In or-
order to vary continuously the electron density in the samples, a metallic gate was evaporated on the surface. In this way intersubband absorption spectra were measured in the carrier-concentration range \( n \sim 2 \times 10^9 - 10^{12} \) cm\(^{-2}\) and in the temperature range 5–300 K. The 2DEG density was carefully determined by Shubnikov-de Haas and Hall measurements as a function of gate voltage \( V_g \). For optical measurements, we fabricated 45°-edge multipass waveguides. Absorption spectra were acquired with an infrared Fourier transform interferometer in normal or step-scan modes and using infrared light polarized perpendicularly to the QW layers and propagating under the gate. In the step-scan case, the bias voltage was square-wave modulated between -2V and the desired voltage, at a frequency between 1 and 20 kHz. In both cases, background spectra were collected at \( V_g = -2 \) V, at this bias the QWs were fully depleted.

Figure 1 shows representative absorption spectra for the 8.7-nm- (left panel) and 7.5-nm-QW structures (right panel) out of a large set taken at many gate-bias values and different temperatures. Dashed lines refer to electron density \( n \sim 8.4 \times 10^{11} \) cm\(^{-2}\) (\( V_g = 0.1 \) V and 0.4 V in the 8.7 and 7.5 nm QWs, respectively), solid lines to \( n \sim 4 \times 10^{10} \) cm\(^{-2}\) (\( V_g = -1.2 \) V, left panel) and \( n \sim 1 \times 10^{10} \) cm\(^{-2}\) (\( V_g = -1.1 \) V, right panel). Data correspond to transitions between the first two subbands in the well, as depicted in the inset of Fig. 1. At the higher electron densities, spectra have a lorentzian line-shape with a full width at half maximum (FWHM) of 3.5 and 4.5 meV. Surprisingly, the measured peak-energy shift at the different biases in Fig. 1 is very small, despite the very different electric fields (up to \( 8 \times 10^4 \) V/cm at the lower carrier densities) and carrier concentrations. Indeed, a single-particle description for the IS energy that includes only the static Hartree term (and therefore takes into account exclusively the appropriate band-bending of the biased heterostructures) does lead to the prediction of a significant energy shift (see Fig. 1 dashed line for the case of the 8.7 nm QW). The observed weak dependence of IS transition energy on electron density is the result of a cancellation effect between Stark shift and dynamic many-body contributions. This is clearly shown in Fig. 2 where measured IS peak energies for the 8.7 nm structure (dots) are plotted as a function of carrier sheet density together with results of a theoretical model (solid line) that includes many-body contributions within the adiabatic local density approximation (LDA) of the density functional theory, and therefore explicitly takes into account the effects of depolarization and excitonic shifts. The IS peak energy was calculated by evaluating the maximum of:

\[
\Re \tilde{\sigma}_{zz}(\omega) \propto \Im m \frac{\omega G_{12}(\omega)}{1 + (\alpha(\omega) - \beta) G_{12}(\omega)}, \tag{1}
\]

where \( \tilde{\sigma}_{zz}(\omega) \) is the \( zz \) component of the frequency dependent conductivity tensor, and \( G_{12} \) is proportional to the response function of a non-interacting 2DEG including the specific energy dispersions of the two individual subbands considered. These are evaluated extending the calculations reported in Ref. 14 and considering the band bending in the entire heterostructure (see also Ref. 1); in all the theory different non-parabolic dispersions in every layer were considered in (1) the depolarization contribution is linked to

\[
\alpha(\omega) = 2\varepsilon^2 n \int \frac{dz'}{\varepsilon(z', \omega)} \left[ \int_0^{z'} dz \xi_2(z) \xi_1(z) \right]^2 \frac{1}{\xi_{12}^2}, \tag{2}
\]

where \( \varepsilon(z, \omega) \) is the layer-dependent dielectric function, whose frequency dependence is estimated like in Ref. 15, \( \xi_{1,2} \) are the wave-functions along the growth direction and \( \xi_{12} \) is the difference between the minima of the two subbands, as calculated from a Schrödinger type equation derived in the LDA formulation of density functional theory. The excitonic contribution is linked to

\[
\beta = -2n \int dz \xi_2(z)^2 \xi_1(z)^2 \frac{d\nu_{xc}(N)}{dN} \frac{1}{\xi_{12}^2}, \tag{3}
\]

where \( \nu_{xc}(N) \) is the exchange-correlation potential as a function of the position-dependent three-dimensional electron density \( N \) determined self-consistently.

The inclusion of all these effects was necessary to obtain the successful description of the experimental results in the broad range of carrier density that is shown in Fig. 2. In particular, the accuracy of our model in determining the 2DEG density is shown in the inset of Fig. 2 where the carrier density estimated by Hall measurements (short-dash line), Shubnikov-de Haas oscillations (empty circles), and area under the absorbance peak (with oscillator strength calculated as in Ref. 1) is reported as a function of \( V_g \) together with the theoretical dependence as derived within LDA (solid line). Figure 2 shows also an excellent agreement between experimental and theoretical results for IS transition energies in a wide range of electron density (similar results were obtained for the second sample). This agreement however is not present at low densities where the theory is not able to reproduce the softening of the IS transition energy. This behavior signals a change in the nature of the intersubband transition and originates from an interplay between many-body effects and disorder. This is the focus of this paper and in what follows we shall discuss the manifestations of this transition and its physical meaning.

In order to analyze the low-electron density behavior of IS transitions, it is useful to refer to Fig. 3 where the deviation of the experimental data from the calculated values is reported for both samples (open squares, right scale) at \( T = 5 \) K. At high density this deviation is \( \sim 0 \) meV for both samples, but for \( n < 3 \times 10^{11} \) cm\(^{-2}\) (8.7 nm sample, upper panel) and \( n < 4 \times 10^{10} \) cm\(^{-2}\) (7.5 nm sample, lower panel) we observe a notable drop in energy of about 2 meV and 3.5 meV, respectively. This evolution was monitored down to very low densities (\( 1.4 \times 10^{10} \) cm\(^{-2}\) and \( 2 \times 10^9 \) cm\(^{-2}\), respectively). The pronounced decrease of the intersubband absorption spectral...
weight hindered measurements at even lower densities. Remarkably, the softening of the IS transition energies occurs together with a rather sudden increase of the corresponding FWHM values. This is also shown in Fig. 2 (solid circles, left scale) where experimental FWHMs are plotted as a function of electron density. In the high density regime the FWHM stays close to a quite small value (around 4 meV). This type of behavior is consistent with calculations that include many-body contributions. In fact it has been shown, and confirmed by our calculation, that these terms bring to its intrinsic homogeneous value, the absorption-peak linewidth drastically reduc-
ing it from the values calculated taking into account the different and non-parabolic dispersions of the subbands, that increase with density. Both the energy position and FWHM trend, therefore, indicate that in the high-density regions absorption originates from purely collective intersubband excitation (charge density excitation – CDE).

The observed behavior at the lower densities signals that a change takes place in the collective response of the 2DEG. This transition occurs when the Coulomb interaction between electrons is not strong enough compared to disorder and cannot concentrate the whole oscillator strength in a narrow CDE mode. In this regime the IS transition incorporates contributions of weakly-interacting randomly localized excitations, and this leads to the observed peak broadening and its shift towards lower energies. This evolution is schematically illustrated in Fig. 3 where the trends in the CDE energy (black line) and in the single particle excitation (SPE) continuum (darker gray areas) are shown as a function of density. The light gray area represents the intrinsic CDE broadening. The darkest gray area includes the non-parabolicity contributions. The dominant source of broadening for single-particle excitations (SPEs), however, is inhomoge-
neous in nature and gives rise to the overall SPE broadening shown in Fig. 4. The observed transition takes place when the broadened CDE and SPE excitations merge. At this density new relaxation channels of the intersubband CDE into localized single-particle modes open, a phenomenon similar to conventional Landau damping at finite in-plane wavevectors. At even lower densities intersubband spectra are characterized by localized SPEs. This second regime was explored by Yakimov et al. thanks to the large fluctuations provided by the self-aggregated quantum dots in the samples they studied. This overall behavior was theoretically discussed in Ref. 1.

It can be noted that the transition region here discussed occurs at higher density for the narrower well, where indeed the electron states are more sensitive to well-width fluctuations. Furthermore, we observed that these changes in energy and linewidth at low densities display a marked temperature dependence and were not detected at temperatures larger than 60 K (data not shown). This suggests an intriguing thermally-induced modification of the collective response of the system. It must be noted in fact that, contrary to this behavior, the many-body contributions to the peak position persisted at all temperatures studied.

However, in order to further corroborate our inter-
pretation, we developed a simple model based on the Drude approach for the response function of a QW characterized by a gaussian-broadened single-particle intersubband transition with a phenomenological inhomogeneous broadening superimposed to a constant homogeneous linewidth. Results for the deviation of the peak position from the depolarization-shifted energy and for the FWHM, as obtained by a lorentzian fit of the calculated response function, are in agreement with the data reported in Fig. 3. Within this simplified model, the transition between the purely collective excitation and the intermediate regime discussed above occurs when the depolarization shift is comparable to the inhomogeneous broadening consistently with our data. Also, the softening of the transition energy occurs because the CDE peak, located in the high-energy end of the broadened single-particle excitation spectrum, possesses all the oscillator strength at high density.

In conclusion, we have experimentally and theoretically studied intersubband transitions in modulation-doped narrow GaAs quantum wells in a wide range of electron densities. We have reported evidence of the damping of the CDE due to coupling with single-particle localized modes as the density is lowered. This transition is a direct manifestation of the influence of the interplay between disorder and many-body effects in determining the nature of IS excitations of the 2DEG in semiconductor heterostructures.

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FIG. 1. Low-temperature quantum well (QW) intersubband absorption spectra at high (dashed line) and low (solid line) electron density for the two structures studied. Measurements were performed in a 2.6-mm-long and 370-µm-high optical waveguide. Dashed lines correspond to electron density \( n \approx 8.4 \times 10^{11} \text{ cm}^{-2} \). Solid lines correspond to \( n \approx 4 \times 10^{10} \text{ cm}^{-2} \) (left panel), and to \( n \approx 1 \times 10^{10} \text{ cm}^{-2} \) (right panel). Inset: Calculated band diagram for the 8.7 nm QW with no applied bias. Horizontal solid lines correspond to the energy of the bottom of the first two subbands; horizontal dashed line is the chemical potential.

FIG. 2. Low-temperature intersubband absorption peak energies as a function of carrier sheet density for the sample with 8.7 nm well width; experimental data are shown as dots with error bars. Solid line: calculated peak energy including many-body effects within the adiabatic local density approximation of the density functional theory. Dashed line is the Hartree approximation. Inset: electron sheet density as a function of applied bias as deduced from Hall (dotted line), Shubnikov-de Haas (open circles), and optical measurements (squares). Solid line is the result of the theoretical model.

FIG. 3. Full width at half maximum (FWHM, solid circles) of the intersubband absorption peak and deviation of measured peak position from theoretical values (\( \Delta E \), open squares) as a function of electron density.
FIG. 4. Schematic representation of charge-density excitation (CDE) energy (solid line) and intersubband single particle excitation (SPE) continuum (gray and dark gray areas) versus electron sheet density. The light gray area represents the intrinsic CDE broadening. The darker gray area shows the broadening originating from subband dispersions, the gray region the inhomogeneous broadening.