Effects of the exchange instability on collective spin and charge excitations of the two-dimensional electron gas

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We report inelastic light scattering measurements of the elementary excitations of a two-dimensional (2D) electron gas formed in a modulation-doped GaAs single quantum well, when the first excited conduction subband begins to be populated and the sudden renormalization of its energy occurs due to an exchange-driven instability of the 2D system. The Hartree and exchange-correlation terms of the Coulomb interaction, which can be precisely determined from the energies of the charge and spin-density excitations in light scattering spectra, exhibit abrupt changes upon filling of the excited subband. Density-functional calculations of the collective excitations within the time-dependent local spin-density approximation provide a simple explanation for the strong enhancement of the Hartree term observed in the experiment. In contrast, the sudden reduction of exchange-correlation vertex corrections at the instability are not well accounted for by the theory. We discuss this discrepancy and present its peculiar temperature dependence.

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I. INTRODUCTION

High-mobility two-dimensional electron gases (2DEG's) formed in modulation-doped semiconductor quantum well structures are particularly suitable for studies of the many-body behavior of dilute electron systems. A vast variety of new phenomena are a consequence of enhanced exchange interactions and correlation energies in low-dimensional systems.[1] In particular, the theoretical prediction of an exchange-driven instability of very diluted electron systems in quantum wells and its effects on different aspects such as magnetic order, excitonic binding, and band-gap renormalization attracted much attention recently.[2–6] Anomalies in the chemical potential and the compressibility of the electron gas were also found in the low-density regime, where exchange-interaction terms and correlations dominate over the kinetic energy.[4,8] In double quantum wells (DQW's), interlayer correlation effects might also induce other types of instabilities such as a (controversial) bilayer-to-monolayer transition[5] or even lead to ferromagnetic or antiferromagnetic order.[6,7] The use of DQW's to study many-body Coulomb effects in electron gases provides the possibility of suppressing localization effects at low densities, using the electron gas in one well for the complete screening of the disorder potential generated by the ionized donors in the other layer. Here we followed a different approach but based on a similar idea. By raising the Fermi energy in a single quantum well structure we slightly populate the first excited subband, whereas the high electron density in the ground state screens the disorder potential. Recent results obtained with this configuration demonstrated the occurrence of a first-order phase transition, as the second subband becomes abruptly populated,[9] and the existence of a low-density phase of the 2DEG characterized by the appearance of a spin texture in the 2D plane.[10]

In Ref. 9 it has been shown that the observed abrupt renormalization of the intersubband energy due to the incipient filling of an excited subband occurs simultaneously with a jump in its electron density from zero to a finite value of about 5×10^{10} cm^{-2}. In contrast, the density in the ground-state subband increases smoothly with the Fermi level. This discontinuity in the electron density involves a thermodynamical instability of the electron gas driven by intersubband exchange interaction terms. This was confirmed by self-consistent calculations of the subband structure and the electron occupation within the formalism of density-functional theory (DFT).[11] The calculations, however, account for such instability only when the exchange potential is treated exactly.[9,11] As the Fermi level becomes resonant with the bottom of the first-excited subband, the system undergoes a first-order phase transition gaining energy by displacing a macroscopic amount of charge within the quantum well, which leads to a sudden reduction of the intersubband spacing energy.

The purpose of this work is to investigate if there is any influence of the phase transition mentioned before on the magnitude of electron-electron Coulomb interactions. It is

well known that many-body effects in the electron gas manifest themselves in the spectrum of collective spin and charge-density excitations. The information about the intersubband exchange-correlation energy, partly responsible for the occurrence of the instability, can be obtained directly from the frequency shift of the spin-density excitation (SDE) from the intersubband single-particle energy (SPE), the so-called excitonic shift.[12,13] On the other hand, the macroscopic displacement of charge associated with the sudden renormalization of the subband energy is expected to have an impact on the Hartree energy which contributes to the intersubband charge-density excitation (CDE).[12,13] With this in mind, both inelastic light scattering measurements and self-consistent calculations within the local spin-density approximation were carried out in the density range of the thermodynamical instability of the 2DEG.

We point out that the exact-exchange theory also predicted a spontaneous breakdown of the spin symmetry to take place in a narrow range of Fermi levels close to the first-order phase transition. The spin instability is favored by *intrasubband* exchange interactions in the excited subband, as long as its population remains low.[10] Spontaneous spin polarization of the 2D system was observed only for low effective electron temperatures. The measurements of the present work, however, were performed at high laser powers, at which the temperature of the electron gas is increased beyond the point of destruction of any magnetic order. The advantage of using high powers is the reduction of the measurement time, what is important due to the unstable character of the system in the density range of interest.

II. EXPERIMENTAL DETAILS

The sample is a modulation-doped 25-nm wide GaAs single quantum well (SQW) with $Al_{0.33}Ga_{0.67}As$ barriers grown by molecular-beam epitaxy. The growth sequence is given in Ref. 14. At 2 K the electron gas has a mobility of 8×10^5 cm^2/V s and a density of about 6.5×10^{11} cm^{-2}. In order to change the electron density, a dc bias is applied between the 2DEG, contacted from the surface by In alloying, and a metallic back contact. Without bias only the lowest subband is occupied. By raising E_F with the gate voltage, the first-excited subband becomes populated.

The electron densities n_0 and n_1 of the ground and first-excited subbands, respectively, as well as the intersubband energy E_{01} and the Fermi level were determined by fitting the PL spectra with a line shape function. It consists of a Lorentzian or Gaussian peak corresponding to the resonant band-to-band transitions, the steplike 2D density of states of the quantum well and the Fermi distribution. The same kind of fitting function is used for describing the contribution to PL emission arising from optical transitions from the first-excited subband. In this case, however, the function reduces to a single Lorentzian (Gaussian) peak if $E_F<E_{01}$. Inelastic light scattering measurements were performed in backscattering geometry with incident photon energies in resonance with an excitonic transition between high excited states of the SQW. A tunable Ti:sapphire laser was used for excitation and spectra were recorded with optical multichannel detection.

III. CALCULATION METHOD

The calculations have been performed in the local spin-density approximation (LSDA) within the framework of the density-functional theory.[15] The spectrum of excitations can be obtained with the so-called time-dependent local spin-density approximation (TDLSDA); a linear response theory which generalizes the well-known time-dependent Hartree approximation or random phase approximation (RPA).

Within LSDA, the exchange-correlation potential is a functional of electronic densities

$$V_{\rm xc}(z,\sigma)=\frac{\partial}{\partial n^{\sigma}}[(n^{\uparrow}+n^{\downarrow})\varepsilon_{\rm xc}(n^{\uparrow},n^{\downarrow})], \tag{1}$$

where $\sigma,\sigma'=1$ (-1) for spin $\uparrow$ $(\downarrow)$ respectively, and $\varepsilon_{\rm xc}$ $(n^{\uparrow},n^{\downarrow})$ is the exchange-correlation energy per particle, as parametrized by Perdew and Zunger[16] for the unpolarized and polarized homogeneous 3D electron gas and interpolated by von Barth and Hedin[17] for an arbitrary magnetization.

The failure of LSDA to achieve a correct self-consistent electronic structure due to a poor treatment of exchange effects is well known as the gap problem. So we proceed as follows: having as initial information the total density n, the Fermi energy E_F, and the energies of the first two subbands E_0 and E_1 from photoluminescence experiments, we adjust the geometry of the SQW in the calculations to obtain self consistently the electronic structure corresponding to the experimental input parameters. The wave functions obtained in this way are very close to those yielded by an exact-exchange calculation.[18] The parameters to be adjusted are the well width d_w and the asymmetry parameter η. The last one reproduces the change in the well potential shape when the bias is introduced. Without bias ($\eta=1$) the well has the original triangular shape, whereas a value of $\eta=0$ correspond to a symmetric SQW.

The collective excitations of charge and spin were calculated using a generalized time-dependent local spin-density approximation.[7] In this approach, the energy of each mode is obtained from the zeros of the secular determinant of the spin-dependent dielectric tensor, which can be written in terms of the exchange-correlation potential in the subband representation. The corresponding potential matrix elements, as a function of the wave vector q of the excitations, are

$$V^{\sigma,\sigma'}_{ij,i'j'}(q)=\frac{1}{A}\int dz\int dz'\,\phi^{*}_{i\sigma}(z)\phi_{j\sigma}(z)\phi^{*}_{i'\sigma'}(z')\phi_{j'\sigma'}(z')$$
$$\times\{(2\pi e^2/\varepsilon q)\exp(-q|z-z'|)+\delta(z-z')$$
$$\times[K_{\rm xc}(z)+(\sigma+\sigma')J_{\rm xc}(z)+\sigma\sigma' I_{\rm xc}(z)]\}, \tag{2}$$

$\phi_{i\sigma}(z)$ are the self-consistent wave functions that diagonalize the effective one-dimensional LSDA Hamiltonian, after assuming translational invariance along the (x,y) plane (area A), and i,j,i',j' are subband indexes. The term in the second line on the right-hand side of Eq. (2) corresponds to the Hartree contribution, with ε being the dielectric constant of GaAs. The terms in square brackets, which all arise from exchange-correlation (xc) contributions, are given as

$$K_{\rm xc}(z)=\partial^2 E_{\rm xc}/\partial n^2|_{n(z),m(z)}, \tag{3}$$

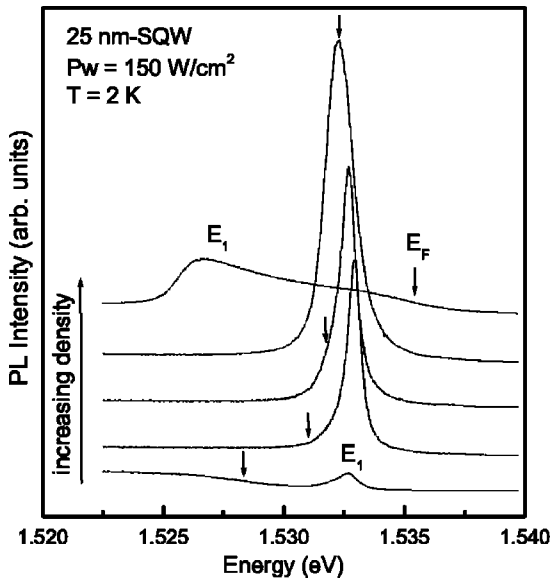

FIG. 1. Photoluminescence spectra of a 25-nm-wide single quantum well in the energy region of the emission from the first-excited subband at E_1. The spectra were taken at 2 K lattice temperature and for different voltages. The evolution at different voltages of the Fermi energy E_F, obtained by fitting the PL spectra with a line shape function, is indicated with arrows.

$$J_{\mathrm{xc}}(z) = \partial^2 E_{\mathrm{xc}}/\partial n \partial m|_{n(z),m(z)}, \tag{4}$$

$$I_{\mathrm{xc}}(z) = \partial^2 E_{\mathrm{xc}}/\partial m^2|_{n(z),m(z)}, \tag{5}$$

where $E_{\mathrm{xc}}(n,m)=n\varepsilon_{\mathrm{xc}}(n,m)$ is the exchange-correlation energy per particle of the homogeneous 3D electron gas for arbitrary density $n(z)=n^{\uparrow}+n^{\downarrow}$ and magnetization $m(z)=n^{\uparrow}-n^{\downarrow}$. $J_{\mathrm{xc}}(z)$ corresponds to a mixed charge-magnetic excitation, whose contribution is zero in non-spin-polarized (paramagnetic) phases, as are the ones studied in this work. In this paramagnetic situation both CDE and SDE are decoupled. In addition to, by using the symmetry $\phi_{i\sigma}(z)=\phi_{i,-\sigma}(z)$ it can be derived from Eq. (2) that the CDE has contributions from the Hartree term and the exchange one proportional to $K_{\mathrm{xc}}(z)$, whereas only the term proportional to $I_{\mathrm{xc}}(z)$ contributes to the SDE. It is worth noting that our calculation includes consistently exchange-correlation effects in both the ground-state calculations and in the linear-response treatment of the excited states. Exchange and correlation is included at the LDA level, as there not exist to date a dynamic (time-dependent) extension of the static exact-exchange results of Refs. 9,11.

IV. RESULTS AND DISCUSSION

Figure 1 shows photoluminescence (PL) spectra of the 2DEG measured at bias conditions of instability at 2 K lattice temperature and a high laser power of about 150 W/cm^2. The peak labeled E_1 corresponds to the optical transition between the bottom of the first-excited electron subband and the hole ground state, which becomes dipole allowed due to the lack of inversion symmetry of the triangular potential in the doped well. The sudden renormalization of the E_1 energy is observed when the Fermi level comes into resonance with the bottom of this subband.[9]

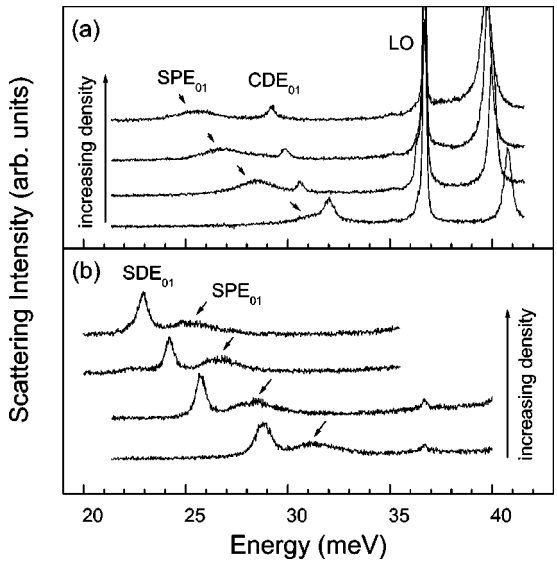

FIG. 2. (a) Polarized (∥) and (b) depolarized (⊥) light scattering spectra of intersubband excitations at different voltages. The peaks corresponding to charge-density (CDE), spin-density (SDE), and single-particle excitations (SPE) as well as the LO phonon are indicated.

In the inelastic light scattering Stokes process, an absorbed photon generates an electron-hole pair that, before recombining, interacts with the system creating an elementary excitation. The energy and momentum difference between the absorbed and emitted photon gives the dispersion relation of the created excitation. The spectrum of the collective modes enables the determination of the direct and exchange-correlation terms of the Coulomb interaction from the energy shifts with respect to the intersubband energy E_{01}. A precise value of the latter can be obtained from the intersubband single-particle excitation (SPE), provided it is observed as broad peak in light scattering spectra. Although the observation of the SPE under extreme resonance conditions is an experimental fact reported for many systems and in different dimensions,[12,19] the reason for it is still not completely understood, since it involves electron-hole pair excitations of the noninteracting electron gas. Clear polarization selection rules are expected for the collective modes. For parallel polarization of the incident and scattered photon should be the charge excitation optically active, whereas the spin-density excitation is forbidden due to spin conservation. The contrary is true for crossed polarization, where spin-flip processes activate depolarized light scattering.

Figure 2 shows inelastic light scattering spectra obtained at 2 K and different voltages in polarized (∥) and depolarized (⊥) configuration. In the spectra with parallel polarization, the LO phonon at about 36.5 meV and the CDE associated with the intersubband transition between the ground-state and the first-excited subband $0\rightarrow 1$ are apparent. The SDE_{01}, in contrast, is observed in the depolarized configuration. The broader peak which appears in spectra with both polarizations is the one assigned to the single-particle excitation

SPE_{01} and is thus centered at the intersubband energy E_{01}. The marked shift of the SDE to lower energies from the intersubband energy demonstrates that exchange vertex corrections are indeed significant in this electronic system. The set of spectra in Fig. 2 illustrates the evolution of the excitation energies by varying the density of the electron gas. The measurements were done under the same conditions of temperature and laser power as for the corresponding PL spectra, some of which are shown in Fig. 1. The shift of the SPE to lower energies reproduces the renormalization of E_1, as observed in PL. Furthermore, the changes of the energy separation between the excitations indicate that the many-body corrections depend on electron density as well.

Within the formalism of TDLSDA the many-body contributions γ_{CDE} and γ_{SDE} to the energies of the charge and spin-density excitation, respectively, can be written in terms of the Hartree (α) and exchange-correlation terms (β_{CDE}, β_{SDE}) of the Coulomb interaction as[12,13]

$$\gamma_{CDE} = \frac{\alpha}{\tilde{\epsilon}(\omega_{CDE})} - \beta_{CDE}, \tag{6}$$

$$\gamma_{SDE} = -\beta_{SDE}, \tag{7}$$

where α represents the depolarization shift, arising from the macroscopic electric field induced by the charge fluctuations themselves. The parameters β_{CDE} and β_{SDE} are the excitonic shifts for conserving and nonconserving spin intersubband transitions, respectively, and $\tilde{\epsilon}(\omega)=(\omega^2-\omega_{LO}^2)/(\omega^2-\omega_{TO}^2)$ is the contribution of the polar lattice to the dielectric function.[20] The depolarization shift α is a positive contribution that typically dominates over the negative one of the excitonic shift in the high-density regime. The exchange-correlation parameter β_i is a function of $\partial V/\partial\rho_i$, where V is the exchange-correlation potential of the electron gas and $\rho_i=n(z)$, $m(z)$ is either the total charge or magnetization. The difference between β_{CDE} and β_{SDE} is a pure correlation term, often neglected in the literature.[21] Here both terms are considered independently, as it will be justified below.

The parameters α and β_i can be calculated from the energy of the collective excitations in the long wavelength limit ($q\approx 0$ for backscattering geometry) according to[21]

$$(\hbar\omega_{CDE})^2 - E_{01}^2 = 2\delta n_{01}E_{01}\left(\frac{\alpha}{\tilde{\epsilon}} - \beta_{CDE}\right), \tag{8}$$

$$(\hbar\omega_{SDE})^2 - E_{01}^2 = -2\delta n_{01}E_{01}\beta_{SDE}, \tag{9}$$

where $\delta n_{01}=n_0-n_1$ is the difference of the subband densities. Thus, from Eqs. (6)–(9) and using the energies of the excitations measured in inelastic light scattering we obtain the values of the parameters γ_{CDE} and γ_{SDE} plotted in Fig. 3 (data points) as a function of the total electron density of the SQW. For comparison the curves represent the theoretical results for the excitation energies yielded by the TDLSDA calculations. The fitting parameters used to reproduce the experimental situations at $T=2$ K for different bias are the asymmetry parameter $\eta=1$, 0.9, 0.87, 0.85, 0.72, 0.34, 0 and a well width $d_w=22.6$ nm. The main result of this work concerns the abrupt change in the many-body corrections to the energies of the elementary excitations occurring at the instability. In the region of the phase transition, i.e., when the Fermi level is almost resonant with the bottom of the first-excited subband, the parameter γ_{CDE} jumps up from almost zero to a finite value, as displayed in Fig. 3. For the parameter γ_{SDE}, in contrast, an abrupt reduction takes place but this cannot be appreciated on the scale of Fig. 3. We discuss this result in detail further below.

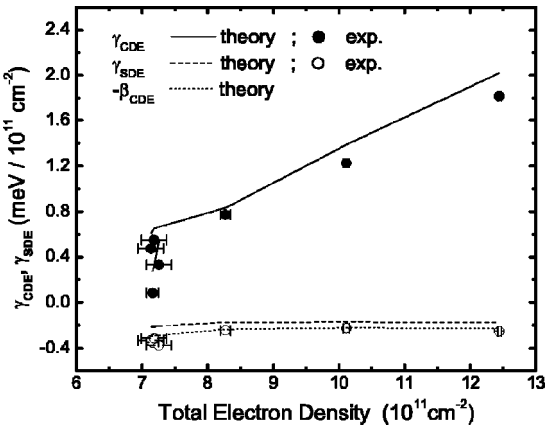

FIG. 3. Intersubband many-body corrections γ_{CDE} and γ_{SDE} to the energies of the respective collective excitation as a function of the total electron density n. Data points were obtained from the peak-energy positions of SDE, CDE, and SPE in light scattering spectra at 2 K lattice temperature and 150 W/cm^2 laser power. The lines correspond to the results of TDLSD calculations.

The abrupt increase of γ_{CDE} when the first-excited subband becomes populated is well accounted for by the TDLSDA calculations. Furthermore, the theoretical results indicate that this behavior has to be ascribed to the Hartree term α. The parameters α and β_{CDE} in γ_{CDE} are calculated independently showing that the main contribution to the jump comes from α, whereas β_{CDE} is small in absolute value. Moreover, the ratio of exchange-correlation terms β_{CDE}/β_{SDE} changes only slightly from 0.71 to 0.79 in the range of total electron densities of Fig. 3. Because of that, we can expect for the experimental results that no significant contribution to the jump in γ_{CDE} stems from exchange and correlation. By inspection of the kernel of the integrals in Eq. (2) related to the Hartree term of the Coulomb interaction we infer that the jump in the depolarization shift α is caused by the abrupt change of the electronic structure and the displacement of a macroscopic amount of charge in real space in the z direction. Because of the triangular shape of the SQW potential, the center of gravity of the wave function in the ground state is shifted with respect to that of the first-excited subband. Hence, the sudden transfer of a net charge from the ground to the first-excited subband leads to a reinforcement of the depolarization electric field associated with α and to a consequent hardening of the intersubband charge-density excitation.

We now turn to the discussion of the intersubband exchange-correlation interaction terms. Figure 4 shows the range of values of γ_{SDE} in more detail. Here the shortcomings of the local-density approximation at the treatment of exchange and correlation become evident. Nevertheless, we

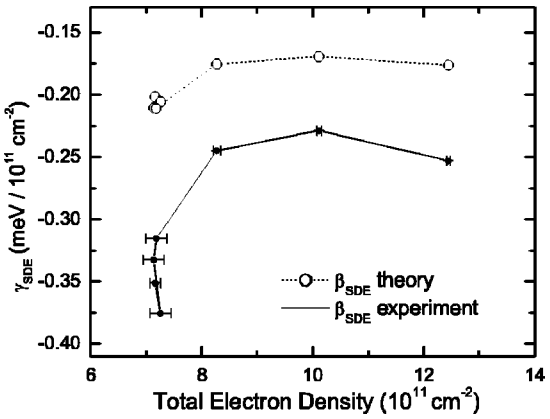

FIG. 4. Exchange-correlation energy correction γ_{SDE} as a function of the total electron density n. Experimental (full points) and TDLSD calculation results (open circles) are shown for comparison.

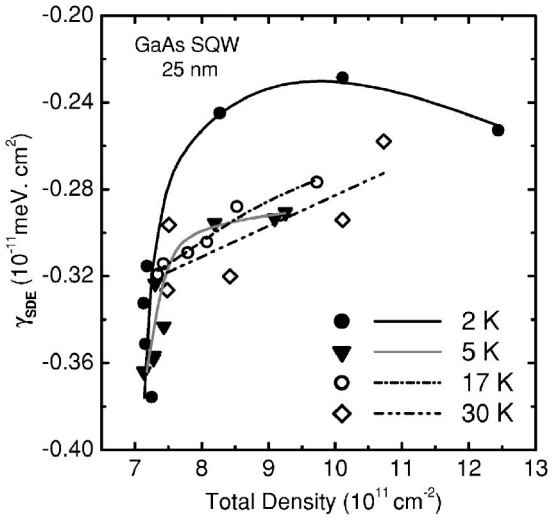

FIG. 5. Evolution with temperature of the parameter γ_{SDE} as a function of the total density. Lines are guides to the eye.

learn much about the many-body behavior of the 2DEG by comparing theory and experiment. There are two points which are worth to be emphasized: (i) the calculated absolute values of γ_{SDE} are about 70% smaller than the measured ones and (ii) the calculations are unable to reproduce the abrupt reduction of the excitonic shift observed at incipient occupations of the excited subband. Strictly speaking, the intersubband excitonic shift γ_{SDE} corresponds to the difference in exchange-correlation self-energies between electronic states of the involved subbands. The *overestimation* of exchange effects across a gap within LDA is well known,[22] in which the exchange (as well as correlation) potentials are computed using results obtained for the gapless, homogeneous 3D electron gas. In our case, LDA overestimates the contribution for the first excited subband, hence yielding smaller absolute values for γ_{SDE}, as seen in Fig. 4. Moreover, within LDA the wave functions and thus the exchange terms are smoothly varying functions of the electron density, making the approximation unable to reproduce an exchange instability. It is important to note that in previous work[9] the inclusion of the *exact* exchange potential for the 2DEG in the calculations was necessary to account for the first-order phase transition observed by photoluminescence means. A similar extension of the theory for the calculation of the excitations exceeds the scope of this work.

Preliminary evidence for the reduction in absolute value of the intersubband exchange-correlation vertex correction by populating excited states was reported before in our work on SQW's.[23] Such a collapse has been also observed in double quantum well structures.[24] As mentioned before, the intersubband parameter γ_{SDE} is the difference of exchange self-energies between first and second subband. Thus, when the excited subband becomes occupied, there must be a partial cancellation of exchange self-energies leading to the sudden suppression of the excitonic shift associated with the SDE. It is instructive to compare with the case of a DQW when the Fermi level is tuned across the symmetric-antisymmetric gap (Δ_{SAS}). In the work of Decca *et al.*,[24] a strong softening of the spin-density mode is observed to occur at substantial occupation of the antisymmetric state with a Fermi energy of about four times Δ_{SAS}. For DQW's the self-energies of both subbands become comparable and they cancel out each other almost completely. The striking result of our work is that the reduction of γ_{SDE} occurs *during* the transition associated with the exchange instability, when the electron density of the upper subband is at most one order of magnitude lower than that of the ground-state one. This speaks for an unexpected enhancement of exchange-correlation interactions in very dilute 2D gases in modulation-doped single quantum wells. The fact that the collapse of γ_{SDE} is not complete is a consequence of the asymmetry of the triangular SQW potential. For our sample γ_{SDE} saturates at a value of about -0.25 meV$/10^{11}$ cm^{-2} in the high-density limit with two largely occupied subbands. We point out that for double-layer systems a spin-density instability of the electron gas was also predicted to occur at very low densities[25] but which could not be confirmed experimentally.[26]

Accounting for the first-order character of the exchange-driven phase transition, we have studied the dependence on temperature of the observed changes in the excitonic shift γ_{SDE} of the intersubband spin-density excitation. The corresponding results for four different lattice temperatures ranging from 2 to 30 K are shown as an example in Fig. 5. The sudden reduction of γ_{SDE} at the density of the transition is still present at 5 K but disappears at higher temperatures, for which γ_{SDE} decreases continuously while the upper subband is being filled. From all the temperature data we can determine a critical temperature of about $T_c = 20$ K for the behavior of the excitonic shift γ_{SDE}. This value agrees reasonably well with that of 35 K obtained from PL experiments for the first-order phase transition of the electron gas upon occupation of the first-excited subband.[9] This is not surprising if we consider that both phenomena are manifestations of intersubband exchange interaction terms.

V. CONCLUSIONS

In conclusion, we have shown that the thermodynamical instability of the 2DEG occurring upon occupation of an

excited subband has strong impact on the many-body corrections to the frequencies of the elementary excitations of the charge and spin density. On the one hand, there is a *geometrical* effect showing up in the magnitude of the depolarization shift α, which is related to the self-consistent change in the electrostatic potential of the doped well due to the sudden transfer of a macroscopic amount of charge from the ground-state to the first excited subband at the first-order phase transition. This leads to the abrupt increase of the Hartree term α, as observed experimentally by inelastic light scattering and nicely reproduced by the TDLSDA calculations. On the other hand, we have obtained direct evidence for an enhancement of exchange and correlation interactions tightly connected to the exchange instability. This manifests itself in an abrupt reduction of the intersubband exchange-correlation vertex correction γ_{SDE} to the energy of the spin-density excitation provided that the lattice temperature is kept below a critical value of about $T_c = 20$ K. Above that temperature γ_{SDE} evolves in a continuous manner with increasing Fermi energy across the transition of the 2DEG. We point out that the theoretical description of such striking behavior of excitonic shifts in dilute 2D electron systems exceeds the capabilities of LDA. The inclusion of the exact exchange potential for the 2DEG in the calculations of the excitations might be the key of success to tackle this intriguing many-body problem.

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