Collective excitations in spontaneously spin-polarized phases of semiconductor double-quantum-well systems

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The intersubband collective modes of the spontaneously spin-polarized phases of semiconductor double-quantum-well systems have been analyzed, generalizing the time-dependent local-spin-density approximation for a consistent treatment of exchange-correlation effects in spin-polarized phases. Abrupt changes in the charge- and spin-density-related collective modes are found as the system passes from unpolarized to polarized ground states through first-order transitions. Excellent agreement with available experimental data is found if the tunneling-induced gap is considered as an adjustable parameter. A suggestion is made that for a tunneling-induced gap of about 0.35 meV the polarized phases should be observable at experimentally accessible densities.

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Semiconductor nanostructures have provided the possibility to control and design electron gases.[1] Accordingly, they are an ideal ground to test fundamental concepts of quantum mechanics and many-body theory. Particularly attractive are double-quantum-well (DQW) systems, because calculations in the local-spin-density approximation (LSDA) of density-functional theory (DFT) show that the ground state can have spontaneous spin-polarized phases (SSPPs), even in absence of a magnetic field.[2] Because the use of DFT and its approximations covers most condensed matter physics,[3] the experimental verification of this fundamental prediction about the electron gas concerns a wide broad public. The issue of SSPPs in two-dimensional electron gases (2DEGs) has received considerable impetus as a possible link between the metal-insulator transition in semiconductor 2DEGs, and a ferromagnetic instability has been suggested quite recently.[4] In a previous work we analyzed the collective modes of the *paramagnetic* (*P*) phase of a GaAs/Al_xGa_{1-x}As DQW system.[5] It was found that three uncoupled modes coexist in the *P* phase: two (one intrasubband, one intersubband) corresponding to charge-density excitations (CDEs), the third being an intersubband spin-density excitation (SDE).

In this paper we provide key information of the experimental detection of SSPPs: we describe the collective modes of the phases where the spin symmetry of the system is spontaneously broken. The electronic excitations in these phases have remained essentially unexplored.[6] As usual in DFT treatments, our basic variable will be the spin-resolved three-dimensional (3D) density $n^{\sigma}(z)$, with σ denoting the spin ($\uparrow$ or $\downarrow$), and z defined as the DQW growth direction (see Fig. 1). For the study of the collective modes in the SSPP we have generalized the so-called time-dependent local-spin-density approximation[3] (TDLSDA) to the case where the system presents a local, finite-density magnetization $m(z) = n^{\uparrow}(z) - n^{\downarrow}(z)$. Previously, we restricted ourselves to the case where $m(z) \equiv 0$, and since only magnetization fluctuations around the *P* ground state were allowed, the SDE and CDE excitations were uncoupled.[5] In contrast, in this work $m(z) \neq 0$, which gives rise to a strong coupling between CDEs and SDEs.

The collective modes can be identified, even in SSPPs, from the zeros of the determinant of the spin-dependent dielectric function ϵ. In the subband representation, the in-plane wave-vector ($\mathbf{q}$) and frequency-dependent (ω) dielectric tensor can be written as

$$\epsilon^{\sigma,\sigma'}_{ij,i'j'}(\mathbf{q},\omega) = \delta_{ii'}\delta_{jj'}\delta_{\sigma\sigma'} - V^{\sigma,\sigma'}_{ij,j'i'}(\mathbf{q})\Pi^{\sigma'}_{i'j'}(\mathbf{q},\omega), \quad (1)$$

where Π^{σ}_{ij} are the spin-dependent polarizabilities[1] of the DQW system, and the potential matrix elements are defined by

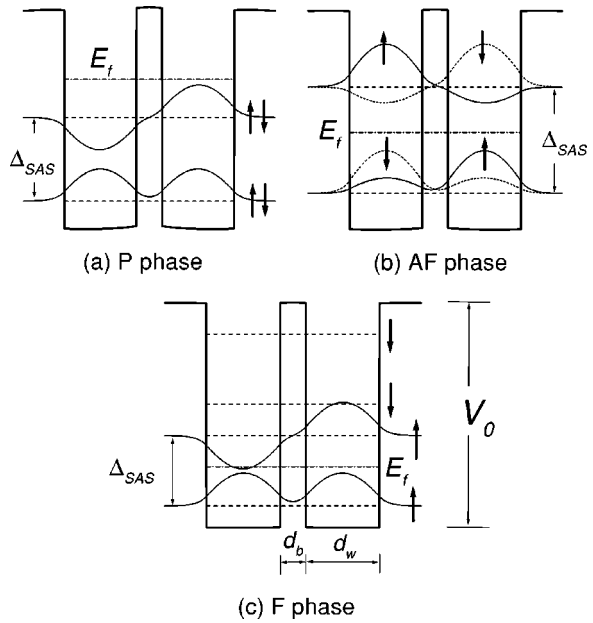

FIG. 1. Schematic drawing (not in scale) of the three possible ground states of the DQW system along the growth direction z. Dashed lines represent the lowest DQW energy subbands, full and dotted lines are the self-consistent wave functions $\phi_{i\sigma}(z)$, and the dash-dotted line is the Fermi energy E_f. V_0 is the bare (undoped) barrier height, and Δ_{SAS} the tunneling-induced gap. Arrows ($\uparrow,\downarrow$) denotes the spin character of each subband wave function.

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

$\phi_{i\sigma}(z)$ 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). i, j, i', and j' are subband indexes, and $\sigma,\sigma'=1$ (-1) for spin $\uparrow$ $(\downarrow)$. 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 the well-acting semiconductor. All terms in the third line are exchange-correlation (xc) contributions. More precisely, $K_{\rm xc}(z)=\partial^2 E_{\rm xc}/\partial n^2|_{n(z),m(z)}$, $J_{\rm xc}(z)=\partial^2 E_{\rm xc}/\partial n\partial m|_{n(z),m(z)}$, and $I_{\rm xc}(z)=\partial^2 E_{\rm xc}/\partial m^2|_{n(z),m(z)}$. $E_{\rm xc}(n,m)=n\varepsilon_{\rm xc}(n,m)$ is the xc energy per particle of the homogeneous 3D electron gas for arbitrary density magnetizations.[7] $J_{\rm xc}(z)$ corresponds to a mixed charge-magnetic excitation, which is zero in the P phase, but makes an important quantitative contribution in the SSPP, giving rise to mode coupling between CDEs and SDEs. It should be noted that this TDLSDA treatment of the DQW system includes, in a *consistent* way, xc effects in both the ground-state calculations and in the linear-response treatment of the excited states; it treats the tunneling between both wells, and gives a *realistic* description of the DQW system through its well and barrier sizes, its barrier height, and its impurity doping profiles.

To provide a physically transparent analysis of the numerical results presented below, we have also performed an analytical study of Eq. (1), by restricting the available subband space to the ground $(i=1)$ and first-excited $(i=2)$ states of the DQW system. Proceeding in this way, and taking the long-wavelength limit $q\to 0$, the eigenmode equation $\det\boldsymbol{\epsilon}=0$ can be solved analytically for $(\hbar\omega)^2$; while the resulting general expression for the modes is rather complicated, it simplifies drastically in the three phases of interest to us: P, antiferromagnetic (AF), and ferromagnetic (F).[8]

Paramagnetic phase: Here $\phi_{i\sigma}(z)=\phi_{i\bar{\sigma}}(z)$; besides, $i=1$ (2) corresponds to the symmetric (antisymmetric) state of the DQW, and $\phi_{i\sigma}(z)=(-1)^{(i+1)}\phi_{i\sigma}(-z)$. These in turn imply that $\Delta^{\uparrow}_{SAS}=\Delta^{\downarrow}_{SAS}\equiv\Delta_{SAS}$, $\Delta n^{\uparrow}=\Delta n^{\downarrow}\equiv\Delta n/2$, and $V^{\uparrow,\uparrow}_{12,12}=V^{\downarrow,\downarrow}_{12,12}$, $V^{\uparrow,\downarrow}_{12,12}=V^{\downarrow,\uparrow}_{12,12}$. $\Delta^{\sigma}_{SAS}=E^{\sigma}_2-E^{\sigma}_1$ is the tunneling-induced gap given by the difference of energy between the first-excited and ground subbands (with the same spin index), while $\Delta n^{\sigma}=n^{\sigma}_1-n^{\sigma}_2>0$ is the occupation density difference between the subbands. Using these symmetries, the eigenvalue equation for the inter-subband collective modes in the long-wavelength limit in the P phase becomes

$$\hbar\omega_{\pm}(q\to 0)\to\Delta_{SAS}\left[1+\frac{\Delta n}{\Delta_{SAS}}(V^{\uparrow,\uparrow}_{12,12}\pm|V^{\uparrow,\downarrow}_{12,12}|)\right]^{1/2}. \quad (3)$$

Antiferromagnetic phase: Here $\phi_{i\sigma}(z)=\phi_{i\bar{\sigma}}(-z)$. For each spin component, the ground state wave functions are essentially localized in opposite wells, this instability being driven by many-body effects. The increment in kinetic energy (along z) is compensated for by an exchange-correlation (mainly exchange) energy gain. The remaining symmetries for Δ^{σ}_{SAS}, Δn^{σ}, and $V^{\sigma,\sigma'}_{12,12}$ are equal to those of the P phase, and accordingly the validity of Eq. (3) extends to the AF phase.

Ferromagnetic phase: Here $\phi_{i\uparrow}(z)=(-1)^{(i+1)}\phi_{i\uparrow}(-z)$, $\Delta^{\uparrow}_{SAS}\equiv\Delta_{SAS}$, $\Delta n^{\uparrow}\equiv\Delta n$, and only $V^{\uparrow,\uparrow}_{12,12}$ contributes in this almost fully spin-polarized phase. In this case Eq. (3) applies once more, under the replacements $\Delta n\to 2\Delta n$ and $V^{\uparrow,\downarrow}_{12,12}\to 0$. It is noteworthy that while in the P and AF phases there are two-intersubband $(\pm)$ modes, just one inter-subband mode is obtained in the F phase. This is because in a fully polarized spin configuration and when $\Delta^{\uparrow}_{SAS}<E^{\downarrow}_1-E^{\uparrow}_1$ the excitations that involve a spin-flip are shifted to higher energies. As a consequence, the modulations of the charge or spin density become the same, and only one mode should be expected here.

In Fig. 2 we display (discrete points) the evolution of the energies of the intersubband excitations covering the ground-state phase diagram in different ways. These full numerical results have been obtained by solving the effective one-dimensional LSDA Hamiltonian using as basis *all* the bounded states of the DQW system (six subbands per spin for this sample), and then finding the roots of $\boldsymbol{\epsilon}(\mathbf{q},\omega)$ for a given $\mathbf{q}$ in this basis.[9] For clarity, we also include a schematic phase diagram typical of the DQW systems.[2]

As N_s decreases [Fig. 2(a)], with N_s being the total 2D density, we found the uncoupled CDE and SDE of the high-density P phase. At a critical density $N_s\simeq 0.33\times 10^{11}$ cm^{-2}, the ground state of the system suffers a $P\to$ AF first-order transition, as the number of occupied subbands (per spin) decreases from 2 to 1. It is important to note that in the P and F phases Δ^{σ}_{SAS} represents the gap between symmetric and antisymmetric states and is mainly a single-particle magnitude. However, when the system enters in the AF phase, Δ^{σ}_{SAS} suffers an abrupt and strong many-body renormalization by about a factor of 2. This explains the high sensitivity of Δ^{σ}_{SAS} to N_s in this phase, contrasted to the essentially independence with N_s of the P and F phases. According to Eq. (3), as both intersubband modes are intrinsically linked to Δ^{σ}_{SAS}, both also suffer an abrupt change at the $P\to$ AF transition. One of these modes crosses the renormalized Δ^{σ}_{SAS} by decreasing N_s, while the lower solution moves essentially following Δ^{σ}_{SAS}, as some sort of generalized SDE, about 0.5 meV below the subband spacing. At a critical density $N_s\simeq 0.12\times 10^{11}$ cm^{-2} the systems becomes P again, this time through a second-order transition. From the collective modes perspective, this continuous transition is signaled by a complete softening of the generalized SDE. As the density decreases further, the system experiments a final $P\to F$ first-order transition, with an abrupt jump toward the unique mode which exists in the F phase, in agreement with Eq. (3) as adapted to the F phase. On the other hand, when we increase the barrier width as shown in Fig. 2(b), we observe how the SDE of the P phase is smoothly softened and that, as soon the system enters in the AF phase, this now generalized SDE appears again and increases its energy up to reach the Δ_{SAS} value for large d_b, well inside the AF phase.

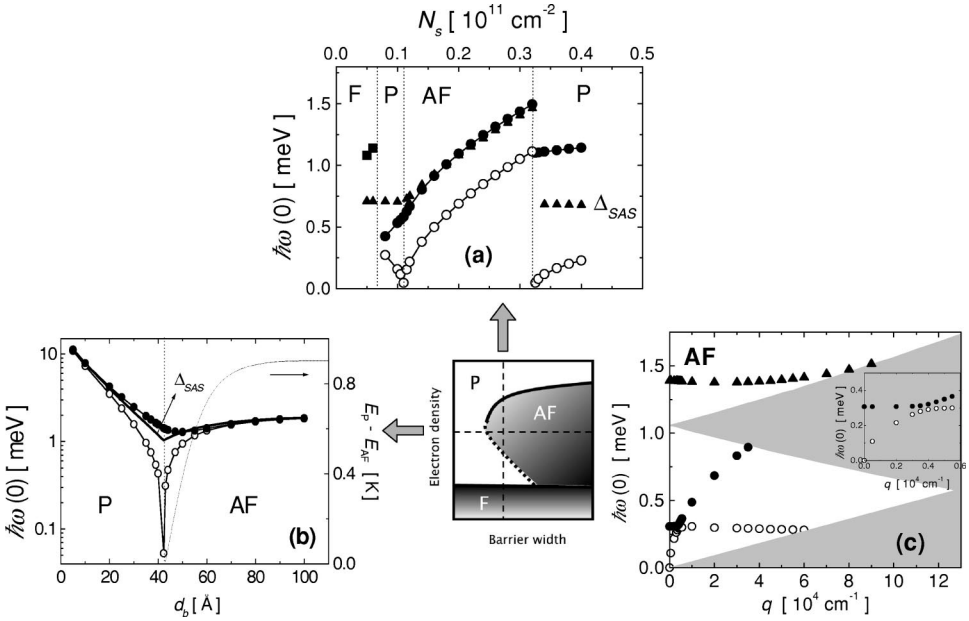

FIG. 2. Collective modes of a GaAs-$Al_xGa_{1-x}As$ DQW system with parameters $d_w = 140$ Å, $V_0 = 220$ meV, and $x \simeq 0.3$. (a) $d_b = 49$ Å, (b) $N_s = 0.25 \times 10^{11}/\text{cm}^2$, and (c) $d_b = 43$ Å, $N_s = 0.25 \times 10^{11}/\text{cm}^2$. Discrete points are the results from the full numerical solution: Full and open circles denote the uncoupled (coupled) CDE and SDE of the P (AF) phase, while full squares represent the unique mode of the F phase. Lines are obtained using Eq. (3), but inserting the self-consistent values for $\Delta^{\sigma}_{SAS}, \Delta n^{\sigma}$, and $V^{\sigma,\sigma'}_{12,12}$ from the full numerical calculation. The dot-dashed line in (b) (right axis) corresponds to the ground-state energy differences between the P and AF phases.

For very narrow barriers in the P phase, Δ_{SAS} increases exponentially. According to Eq. (3) applied to the one subband regime, both the CDE and SDE tend toward Δ_{SAS}; this explains the convergence of both modes toward Δ_{SAS} in this strong tunneling regime. The convergence that occurs for large d_b in the AF phase is simply due to the fact that as the wells are increasingly uncoupled, $V^{\sigma,\sigma'}_{12,12} \to 0$ as the interwell subband overlap decreases. Note the nonintuitive *increase* of Δ_{SAS} with d_b as the system enters in the AF phase; the reason for this is that in this phase an important fraction of Δ_{SAS} is due to many-body effects, which are reinforcing as d_b increases.[2] It is interesting to note that the abrupt jumps in Δ_{SAS} (see Fig. 2) and the associated subband structure (schematically shown in Fig. 1) could be measured by photoluminescence experiments, as in Ref. 10. We also include, in Fig. 2(c), the dispersion relations associated with the three collective modes of the AF phase, close to the boundary between the P and AF phases in Fig. 2(b). The most noticeable feature here is the strong anticrossing between the intrasubband CDE-like and intersubband SDE-like collective excitations, *which is a unique feature of SSPPs*. Because in principle these dispersion relations could be measured, this anticrossing could be used as a complementary tool to confirm the existance of SSPPs, in addition to the long-wavelength jumps displayed in Fig. 2(a) and the softening in Fig. 2(b).

Finally, we test the accuracy of the present calculation by comparison with available experimental data. In Ref. 5 we found a very good agreement with ILS experiments on DQWs with the wells so far apart that $\Delta_{SAS} \simeq 0$ (Ref. 11); in addition, the system was well inside the P high-density regime, where xc effects are small corrections. Here we choose the strongly tunneling-coupled GaAs/$Al_xGa_{1-x}As$ DQW analyzed by Plaut *et al.*[12] In Fig. 3 we draw our results for this system (lines), together with the experimental data (points). It is important to note the weak but discernible minimum which develops on the SDE channel, both in the *theoretical and experimental* curves. While not a complete softening, this feature is a clear indication that the xc-driven instability discussed here is operative. Using as input of the calculation the nominal experimental values for d_b, d_w, and V_0, the agreement between experiment and theory (dotted line) is not good, as the theoretical SDE shows a more pronounced softening than the experimental one. In this case, $\Delta_{SAS} \simeq 1$ meV at the lowest density both experimentally and theoretically. A very good agreement is obtained, however, if we increased Δ_{SAS} to about twice its experimental value ($\simeq 2$ meV), by decreasing d_w from 180 Å to 140 Å (full line).[13] This important quantitative failure could be due to finite temperature and xc effects not included in our zero-

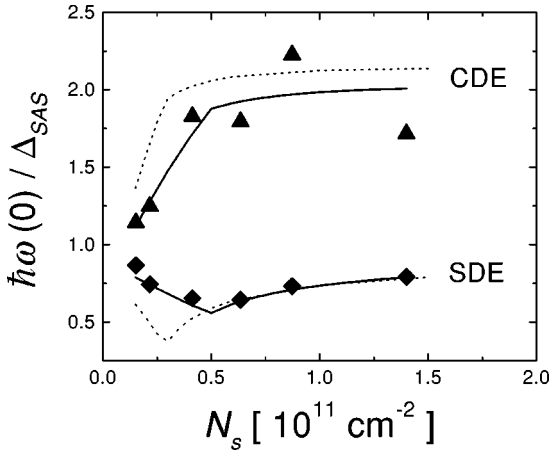

FIG. 3. Symbols: experimental data from Ref. 12, with nominal growth parameters $d_b = 80$ Å, $d_w = 180$ Å, and $V_0 = 45$ meV (Al content $x = 0.1$). Dotted lines, present calculation for the same set of parameters. Full lines: present calculations for $d_w = 140$ Å (same d_b and V_0). For these sets of parameters, the ground state is in the P phase. Note that in this figure lines correspond to the full numerical solution of Eq. (1), without resorting to the analytical result given by Eq. (3).

temperature TDLSDA.[14] From the results presented in Fig. 2(b) for the ground-state energy differences between the *P* and AF phases, one can estimate that the 2DEG should be cooled below 1 K to achieve the instability. Also, the necessity for the adjustment of Δ_{SAS} calls for more accurate treatments of the xc effects in the collective modes beyond LSDA, as in the exact-exchange DFT formalism.[10] It is worth emphasizing, however, that the fundamental physics which drives the DQW system to the SSPP's is unaltered, being an intrinsic feature of these quantum heterostructures. Moreover, and encouraged by the agreement shown in Fig. 3 between theory and experiment by doubling the Δ_{SAS}, we believe that exists the possibility to detect the AF phase in the same range of electronic densities by decreasing the temperature on a sample similar to that studied in Ref. 12, but with a Δ_{SAS} of about 0.35 meV.

In conclusion, we have generalized and applied the TDLSDA to the study of the collective intersubband excitations of the SSPP of semiconductor DQW systems. Abrupt jumps in the long-wavelength limit of the CDE and SDE are found as the system passes from the *P* to AF ground states through a first-order transition. A comparison with available experimental data indicates a *quantitative* failure of the TDLSDA, which has been corrected by adjusting the tunneling-induced gap. This allow us to suggest a feasible experimental configuration suitable to measure these long-sought many-body driven instabilities at zero magnetic field.

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[9] Material parameters: dielectric constant ε ($Al_xGa_{1-x}As$) $\simeq \varepsilon$ (GaAs) $\simeq 10.9$, and effective mass m^* ($Al_xGa_{1-x}As$) $\simeq m^*(GaAs) \simeq 0.067 m_0$ (m_0 being the bare electron mass), neglecting the small differences between well and barrier semiconductors. Essentially the same results are obtained by using the static value $\varepsilon(GaAs) \simeq 13$. Also, two delta-doped donor distributions have been symmetrically located far from the DQW system.

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[13] It is important to realize here that the relevant parameter is Δ_{SAS}, and that decreasing d_w is just a way to increase its value; an alternative procedure could be decrease the barrier height or width.

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