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Exchange instability of the two-dimensional electron gas in semiconductor quantum wells

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A two-dimensional (2D) electron gas formed in a modulation-doped GaAs/$Al_xGa_{1-x}As$ single quantum well undergoes a first-order transition when the first excited subband is occupied with electrons, as the Fermi level is tuned into resonance with the excited subband by applying a dc voltage. Direct evidence for this effect is obtained from low-temperature photoluminescence spectra that display the sudden renormalization of the intersubband energy E_{01} upon the abrupt occupation of the first excited subband. Calculations within density-functional theory, which treat the 2D exchange potential *exactly*, show that this thermodynamical instability of the electron system is mainly driven by *intersubband* terms of the exchange Coulomb interaction, thus being a unique but fundamental property of an electron system with more than one occupied subband.

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Exchange effects are a fundamental manifestation of electron-electron interactions in many-particle systems such as high-mobility two-dimensional (2D) electron gases that form in modulation-doped semiconductor quantum wells. Exchange-correlation interactions are at the origin of a vast variety of fascinating quantum phenomena,[1] for example, spin excitations, magnetic ordering, excitonic binding and band-gap renormalization, among others. The latter, for instance, is apparent in semiconductor heterostructures as a reduction of gap energies when the corresponding band states become populated with carriers either by varying doping levels[2,3] or under intense photoexcitation.[4] This effect is explained as arising from exchange-correlation corrections due to the presence of free carriers in the system.[5,6]

Electronic correlations dictate the behavior of electron gases in reduced dimensions particularly in high magnetic fields and/or at very low densities. The formation of fractional quantum Hall states exhibiting completely unforeseen physical properties is a paradigmatic example of the importance of correlation effects.[7] At zero magnetic field the occurrence of a metal-insulator transition (MIT) in two dimensions[8,9] is not completely understood in terms of scaling theory, which predicts that a *noninteracting* 2D electron or hole system becomes localized at low temperatures for any degree of disorder.[10] Recently, other thermodynamic properties of 2D electron gases such as the chemical potential and its density derivative, the compressibility, have been found to exhibit anomalous behavior across the metal-insulator transition deviating from what is expected within Hartree-Fock theory.[11] In spite of the universalities detected at the MIT, for example, in the temperature dependence of the resistivity,[12] this transition does not reveal a purely intrinsic property of interactions in low-dimensional systems since disorder plays the crucial role. On the contrary, localization effects can be widely suppressed in double-layer electron gases, in which the electron gas in one quantum well is used for the complete screening of the disorder potential of the ionized donors.[13] Using a capacitance technique[13] it was shown that at low densities but without magnetic field the compressibility of a high-mobility 2D electron gas becomes *negative* owing mainly to exchange interactions. This leads to a thermodynamical instability of the electron system. Furthermore, exchange effects might induce other types of instabilities in double quantum wells such as a (controversial) bilayer-to-monolayer transition[14] or even one towards a magnetic ground state.[15] The exchange interaction also gives rise to first-order transitions in diluted magnetic semiconductors.[16]

In this paper we show that at low temperature and zero magnetic field a 2D electron gas formed in a single GaAs quantum well undergoes a first-order phase transition, as the first excited electron subband becomes populated with electrons by raising the Fermi level with a gate voltage. The evidence is found in the sudden and abrupt renormalization of the energy of the first excited subband, as determined from photoluminescence and inelastic light scattering measurements. Self-consistent density-functional calculations with *exact* exchange potential for a 2D electron system reveal that this transition is driven by *intersubband exchange interactions* that provide a feedback mechanism for charge transfer into the excited subband. This theory further predicts the first-order character of the transition. In fact, experiments performed at different temperatures indicate the existence of a critical point at around (35 ± 5) K.

The sample consists of a modulation-doped 245-Å-wide GaAs single quantum well with $Al_{0.33}Ga_{0.67}As$ barriers grown by molecular-beam epitaxy. The growth sequence is given elsewhere.[9] Without bias only the lowest subband is occupied with electrons with Fermi energy $E_F \approx 25$ meV. The energy separation to the second subband is $E_{01} \approx 28$ meV. The electron gas is contacted from the surface by In alloying in order to apply a dc bias up to 30 V between it and a metallic back contact. Photoluminescence (PL) and inelastic light scattering spectra were excited with a tunable Ti:sapphire laser and recorded with optical multichannel detection.

The electron density in the quantum well increases with applied voltage shifting the Fermi level towards degeneracy

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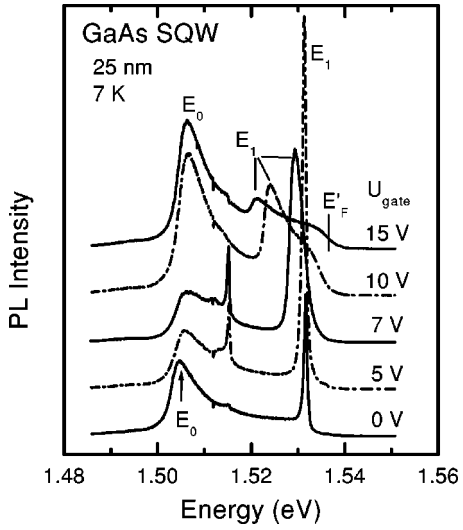

FIG. 1. Photoluminescence spectra of a modulation-doped 25-nm-wide single quantum well at 7 K and for different gate voltages. The position of band-gaps and Fermi energy is indicated. The peak at 1.515 eV arises from bulk GaAs luminescence.

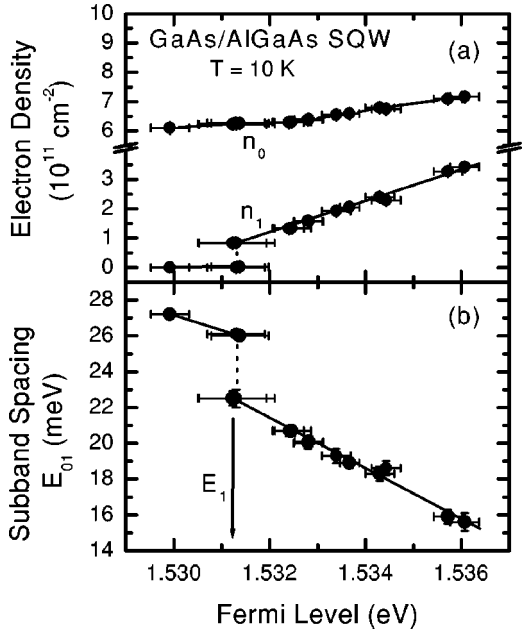

FIG. 2. Dependence on Fermi level of (a) the electron densities n_0 and n_1 of the lowest and the first excited subband, respectively, and (b) of the intersubband spacing E_{01}, as determined from PL spectra at 10 K. The curves are a guide to the eye.

with the bottom of the first excited subband. Figure 1 shows PL spectra recorded at 7 K for different bias. The emission line E_0 corresponds to recombination processes between the lowest electron and heavy-hole subbands (subband index 0). The peak labeled as E_1 is associated with optical transitions between the first excited electron subband and the hole ground state, which become dipole allowed due to the lack of inversion symmetry of the triangular potential in the doped well. The peak at 1.515 eV originates from near band-gap emission of the GaAs buffer layer. At high voltages a high-energy cutoff at the Fermi energy $E_F'=E_F(1+m_e/m_h)$ is clearly apparent from the PL spectra. The factor containing the ratio of electron and hole effective masses accounts for the curvature of the valence band. With increasing bias the Fermi energy increases leading to a population of the first excited subband. An important result concerns the strong redshift of the energy E_1 by about 11 meV. Moreover, the occupation of the second subband proceeds abruptly at the voltage for which the Fermi level reaches its bottom. This reduction of E_1 is a consequence of band-gap renormalization effects in electron gases due to exchange-correlation corrections.[5,6]

The dependence on bias of the electron densities n_0 and n_1 of the ground state and first excited subband, respectively, as well as the intersubband energy E_{01} has been determined from a quantitative analysis of PL line shapes, as described elsewhere.[3,9] The values for n_0, n_1, and E_{01} obtained at 10 K are plotted in Fig. 2 as a function of the Fermi level referred to the top of the valence band. Below 40 K when the Fermi level equals E_1 the electron density n_1 jumps from zero to a finite value ranging from 3×10^{10} to 8×10^{10} cm^{-2} depending on temperature. The electron density of the lowest subband, in contrast, increases slightly but smoothly with voltage. Simultaneously, with the abrupt filling of the second subband, a sudden reduction of the intersubband energy E_{01} by about 3.5 meV is observed.[17] Band-gap renormalization acts as a feedback mechanism for subband filling leading to a sudden population of the excited electron subband and, in addition, causing the pronounced decrease of the intersubband spacing in the well. We point out that PL provides a means for the determination of the Fermi level, which is independent of the quality of the contacts or the way in which charge flows into the quantum well. We, thus, rule out spurious charging effects as cause of the observed jumps in 2D density and intersubband spacing.

The observed discontinuities in electron density and gap renormalization upon occupation of the first excited subband point to a thermodynamical instability of the 2D electron gas. In order to enlighten this fundamental issue, we have performed self-consistent calculations of the subband structure and level occupation of the single quantum well within the formalism of density-functional theory. Our calculation differs from the standard local-density approximation in two major points. The exchange interaction is treated *exactly*[18] for a quasi-2D electron system and the number of particles is *not fixed* but is allowed to change. Mathematically, the exchange potential is calculated according to

$$V_x(\mathbf{r})=\frac{\delta E_x}{\delta\rho(\mathbf{r})}$$
$$=\int \mathbf{dr}'\left\{\sum_{\mathbf{vk}}\int \mathbf{dr}''\left[\frac{\delta E_x}{\delta\phi_{v\mathbf{k}}(\mathbf{r}')}\frac{\delta\phi_{v\mathbf{k}}(\mathbf{r}')}{\delta V_{KS}(\mathbf{r}'')}+\text{c.c.}\right]\right.$$
$$\left.+\sum_v\left[\frac{\delta E_x}{\delta k_F^v}\frac{\delta k_F^v}{\delta V_{KS}(\mathbf{r}'')}\right]\right\}\frac{\delta V_{KS}(\mathbf{r}'')}{\delta\rho(\mathbf{r})}, \qquad (1)$$

where E_x is the exact exchange energy, $\rho(\mathbf{r})$ is the electron density, $V_{KS}(\mathbf{r})=V_{ex}(\mathbf{r})+V_H(\mathbf{r})+V_x(\mathbf{r})+V_c(\mathbf{r})$ is the

Kohn-Sham potential given as sum of the external, Hartree, exchange, and correlation potentials, respectively, and $\phi_{n\mathbf{k}}(\mathbf{r}) \propto e^{i\mathbf{k}\cdot\rho} \cdot \xi_n(z)$ is the wave function of an electron in the quantum well characterized by the envelope function $\xi_n(z)$ in the direction of confinement z. The first term in Eq. (1) represents functional derivatives of E_x with respect to the *shape* of the wave functions,[18] whereas, the second one accounts for the variation of the exchange energy with occupation.

Integration in the (x,y) plane yields (in atomic units)

$$V_x(z) = \sum_{vv'n} I_1(v,v',n) \int dz' \frac{\xi_v(z')\xi_n(z')}{\varepsilon_v - \varepsilon_n} \chi^{-1}(z,z') + \sum_{vv'} I_2(v,v') \int dz' |\xi_v(z')|^2 \chi^{-1}(z,z'), \quad (2)$$

$$I_1(v,v',n) = -k_F^v k_F^{v'} \int \frac{d\rho}{\rho} J_1(k_F^v \rho) J_1(k_F^{v'} \rho) \times \int dz_1 dz_2 \frac{\xi_v(z_1)\xi_n(z_2)\xi_{v'}(z_1)\xi_{v'}(z_2)}{\sqrt{\rho^2 + (z_1 - z_2)^2}}, \quad (3)$$

$$I_2(v,v') = \frac{k_F^{v'}}{2} \int d\rho J_0(k_F^v \rho) J_1(k_F^{v'} \rho) \times \int dz_1 dz_2 \frac{\xi_v(z_1)\xi_v(z_2)\xi_{v'}(z_1)\xi_{v'}(z_2)}{\sqrt{\rho^2 + (z_1 - z_2)^2}}, \quad (4)$$

where the operator $\chi(z,z') = \delta\rho(z)/\delta V_{KS}(z')$ is related to the Linhard susceptibility of the 2D gas in the $T=0$ limit. In Eq. (2) v and v' sum only occupied subbands, while the index n runs over all subbands (with exception of $n=v$). $J_i(z)$ stands for the Bessel function of order i, ρ is the in-plane coordinate vector and k_F^v and $k_F^{v'}$ are the Fermi wave vectors of the occupied subbands.

The striking result of our theory is that the exchange potential $V_x(z)$ changes discontinuously every time a subband becomes occupied. For our experimental situation we have $v = v' = 0$, or $v, v' = 0,1$ for one or two occupied subbands, respectively. The crucial point is that the second term in Eq. (2) remains finite even for $k_F^{v=1} \to 0^+$ because $I_2(1,0) \neq 0$, all other contributions involving occupation of the second subband tend to zero in this limit. The finite term $I_2(1,0)$ is an *intersubband* exchange contribution $(v \neq v')$. From Eq. (4) it is clear that the size of this term is determined by the wave-function overlap between the involved subbands. For our single quantum well this overlap is fairly large. Hence, due to strong intersubband exchange terms, it is more favorable for the 2D electron gas to transfer a macroscopic amount of charge into the first excited subband upon occupation leading to the sudden collapse of the subband spacing E_{01}. The self-consistent solution of the Kohn-Sham equations with $V_x(z)$ given by Eq. (2), in fact, exhibits an abrupt jump from the configuration with one subband occupied to that with two occupied subbands when the Fermi energy equals E_{01}. The calculations yield a sudden reduction of E_{01} by about 2.5 meV for an initial value of 26.5 meV at 10 K, which is in good quantitative agreement with the experimental observation. This is a clear signature of an exchange-driven first-order phase transition, a general and robust effect that occurs at relatively high densities and accordingly leads to quite sizable jumps in subband occupancies and spacings. The abrupt transitions reported here are qualitatively different from the ones reported in Ref. 15, where as result of the essentially zero wave-function overlap between ground and excited subbands the physics is dominated by the much weaker *intrasubband* exchange interaction. More details of the theory and the self-consistent calculations will be given elsewhere.[19]

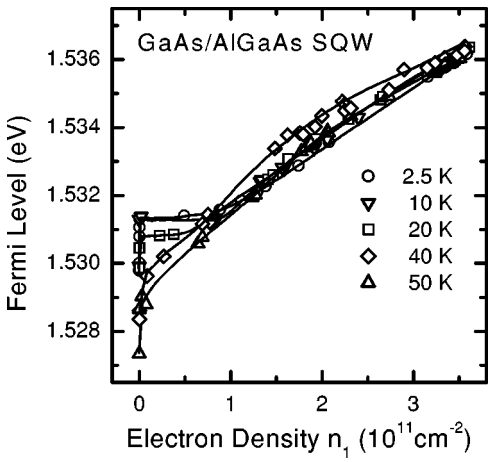

FIG. 3. Measured isothermal curves of Fermi level versus electron density of the first excited subband for the 2D electron gas of the modulation-doped GaAs single quantum well. Above 30 K the discontinuity in n_1 is no longer observed (see text).

Further evidence for the first-order character of the observed exchange instability can be gained from PL measurements at different temperatures. Figure 3 displays several isothermal curves representing the variation of the Fermi level and the density of the first excited subband. The isotherms fall into two classes according as the temperature lies below or above a critical value of $T_c = (35 \pm 5)$ K. Below T_c the 2D electron gas can exist in two states with and without population of the first excited subband, its occupation proceeds abruptly because of the compressibility being negative due to the exchange interactions. Above the critical temperature, on the contrary, the discontinuity in the density n_1 disappears and the system evolves continuously from one phase to another. Additional evidence of a phase mix revealing a spatial inhomogeneity of the electron gas is obtained from luminescence. At the instability and with fixed voltage, PL spectra display a multiple-peak time-varying structure at energies around E_1. Each peak of this feature is assigned to PL emission at E_1 arising from different regions within the laser spot with slightly different electron density.

In conclusion, we have shown experimentally and theoretically for the first time that 2D electron gases formed in a modulation-doped GaAs single quantum well undergo a first-order phase transition when an excited subband be-

comes populated. The signature of this transition is seen in the abrupt renormalization of the subband energy and the related jump in electron density upon occupation of the excited subband. Furthermore, we have determined from the isotherms a critical transition temperature of about 35 K. Self-consistent calculations within density-functional theory, which treat the Coulomb exchange in the 2D system exactly, hence going beyond the state-of-the-art local approximations, show that such thermodynamical instability of the electron gas is mainly induced by intersubband exchange terms. This instability corresponds, for instance, to the same universal class of liquid-vapor phase transitions characterized by being of first order and having a latent heat associated with, which in this case originates from exchange effects. In this way, we have provided further insight into the fundamental issue of the many-body behavior of high-mobility electron gases in 2D arising from exchange interactions between correlated electrons in different subbands of semiconductor quantum wells.

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