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_{x}Ga_{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.

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, 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 or under intense photoexcitation. This effect is explained as arising from exchange-correlation corrections due to the presence of free carriers in the system.

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. At zero magnetic field the occurrence of a metal-insulator transition (MIT) in two dimensions 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. 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. In spite of the universality detected at the MIT, for example, in the temperature dependence of the resistivity, 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. Using a capacitance technique 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 or even one towards a magnetic ground state. The exchange interaction also gives rise to first-order transitions in diluted magnetic semiconductors.

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. 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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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 $E_F$ 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.\textsuperscript{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.\textsuperscript{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 $3 \times 10^{10}$ to $8 \times 10^{10}$ 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.\textsuperscript{11} 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\textsuperscript{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

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

where $E_x$ is the exact exchange energy, $\rho(r)$ is the electron density, $V_{KS}(r) = V_{xc}(r) + V_{HF}(r) + V_{xc}(r) + V_{xc}(r)$ is the
Kohn-Sham potential given as sum of the external, Hartree, exchange, and correlation potentials, respectively, and 
\( \phi_{n}(z)e^{iK_{\mathbf{r}}} \) 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 
\( E_{n} \) with respect to the shape of the wave functions, whereas, 
the second one accounts for the variation of the exchange energy with occupation.

Integration in the \((x,y)\) plane yields (in atomic units)
\[
V_{\mathbf{z}}(z) = \sum_{n} \{ I_{1}(v,v',n) \int d\zeta \frac{\xi_{n}(\zeta')\xi_{n}(\zeta')}{\epsilon_{v}-\epsilon_{n}} \chi^{-1}(z,\zeta') 
+ \sum_{n} I_{2}(v,v') \int d\zeta' \chi^{-1}(z,\zeta') \} 
+ \sum_{n} I_{3}(v,v') \int d\zeta' \chi^{-1}(z,\zeta') \},
\]
where the operator \( \chi(z,\zeta') = \delta \rho(z)/\delta V_{KS}(\zeta') \) 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 \( i \), \( \rho \) is the 
in-plane coordinate vector and \( k_{F}^{v} \) and \( k_{F}^{v'} \) are the Fermi wave 
pression for the 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 
especially 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

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 
inescence. 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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17 We note that the same dependence of \( E_{10} \) on bias has been determined from inelastic light scattering by elementary excitations of the 2D gas associated with electronic transitions between subband 0 and 1.