Evidence of breakdown of the spin symmetry in diluted 2D electron gases

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Abstract – Direct evidence of spin polarisation in dilute two-dimensional electron gases formed in modulation-doped single quantum wells has been obtained from resonant inelastic light scattering. The abrupt enhancement of the exchange-correlation energy of collective intersubband spin and charge excitations observed at very low occupations of the second subband is the signature of the breakdown of the spin symmetry. Calculations of the elementary excitations within the time-dependent local spin-density approximation provide an explanation for the striking behaviour of the different terms of the Coulomb interaction and predict the existence of a ferromagnetic ground state in the very diluted regime.

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Introduction. – Long standing theoretical discussions of spontaneous spin polarisation in dilute electron gases [1–5] were revived by recent experimental claims [6–8]. Weak ferromagnetism observed few years ago in La-doped calcium hexaboride was tentatively explained in terms of a partially polarised three-dimensional fluid [2]. Moreover, last year possible ferromagnetism was claimed to be observed in transport experiments on a GaAs single quantum well (SQW) with a single occupied subband [7]. This evidence, though, is not conclusive due to the effects of localisation. In two dimensions, the stabilisation of a ferromagnetic fluid at low densities might be hindered by the occurrence of the metal-insulator transition [9]. To circumvent localisation in the two-dimensional electron gas (2DEG), we investigated the low populated second electron subband, where the disorder potential is mainly screened by the high density of the first subband.

Recently, we have shown experimentally as well as theoretically that at low temperatures and zero magnetic field a 2D electron gas realized in a GaAs single quantum well (SQW) undergoes a first-order phase transition, as a result of intersubband exchange-correlation terms when the second electron subband becomes populated [8]. The situation of two occupied subbands resembles that of double-layer structures, a system with a rich ground-state phase diagram, where a collapse of the normal metallic state towards a spontaneous magnetic order at very low densities was also predicted from ground-state [3] and collective excitation calculations [4]. Evidence of this first-order phase transition is the abrupt renormalisation of the second subband energy and its sudden population with electrons [10], as observed by photoluminescence (PL) spectra. The first-order character of the transition was confirmed by self-consistent density-functional calculations, including exchange interaction exactly [11,12]. The exact-exchange theory predicted also a spontaneous breakdown of the spin symmetry to take place for a low population of the second subband. In this situation, a new phase was found in PL spectra at low temperatures and at zero magnetic field [8]. The behaviour of this phase under
variation of density, temperature and external magnetic field speaks for the formation of spin-polarised domains with different in-plane magnetisation, as expected for planar ferromagnets to minimise the stray field. However, a direct evidence of the existence of a ferromagnetic phase is still lacking and the interpretation of the PL results remains speculative.

This letter reports on the breakdown of the spin symmetry in a 2DEG, revealed by the abrupt enhancement of the exchange and correlation terms of the Coulomb interaction, as determined by inelastic light scattering from the energies of the collective charge and spin excitations occurring at very low population of the second subband. Calculations within the time-dependent local spin-density approximation (TDLSDA) support the experimental evidence for the existence of a ferromagnetic ground state in the diluted regime.

**Experimental details.** – The 2DEG forms in modulation-doped GaAs/Al<sub>0.33</sub>Ga<sub>0.67</sub>As single quantum wells of 20, 25 and 30 nm well thickness grown by molecular-beam epitaxy. The 2DEG has a mean mobility of about 8×10<sup>5</sup> cm<sup>2</sup>/Vs and a density such that only the lowest subband is occupied with Fermi energies E<sub>F</sub> between 25 and 28 meV at 4.2 K. The electron density can be changed by applying a dc bias between the 2DEG and a metallic back contact. Photoluminescence and inelastic light scattering spectra were obtained using low power densities in the range from 0.5 to 5 W/cm<sup>2</sup>. Light scattering spectra were recorded in backscattering geometry using a line focus in order to avoid heating of the 2DEG at the required laser powers.

The set of spectra in fig. 1 illustrates the evolution of the energy of the charge (CDE), spin (SDE) and single-particle excitations (SPE), associated with the inter-subband transitions 0→1 and 1→2, when varying the density of the electron gas. The shift of the SPE<sub>01</sub> to lower energies and of the SPE<sub>12</sub> to higher energies are consequences of the renormalisation of the second subband with increasing occupation. Furthermore, the change in the energy separation between excitations indicates a dependence of the many-body corrections on electron density [13,14].

**Calculations.** – The ground-state calculations have been performed in the local spin-density approximation (LSDA) in the framework of density-functional theory. We adjusted the geometry of the SQW in order to reproduce the Fermi energy E<sub>F</sub>, and the two subband energies E<sub>0</sub> and E<sub>1</sub> as obtained from photoluminescence experiments.

The energy of the collective excitations of charge (CDE) and spin (SDE) were obtained with TDLSDA from the zeros of the secular determinant of the spin-dependent dielectric tensor, which can be written in terms of the Hartree (V<sub>H</sub>) and exchange-correlation potentials (V<sub>xc</sub>) in the subband representation. The corresponding potential matrix elements, as a function of the wave vector q of the excitations, are

\[
(V_H)_{ij,i'j'}^{\sigma\sigma'}(q) = \frac{1}{A} \int d\mathbf{z} d\mathbf{z'} (2\pi\varepsilon^2/q) \exp(-q|z-z'|) \phi_{\sigma}(z) \phi_{j\sigma}(z) \phi_{i'\sigma'}(z') \phi_{j'\sigma'}(z'),
\]

(1)

\[
(V_{xc})_{ij,i'j'}^{\sigma\sigma'}(q) = \frac{1}{A} \int d\mathbf{z} \phi_{\sigma}(z) \phi_{j\sigma}(z) \phi_{i'\sigma'}(z) \phi_{j'\sigma'}(z) \times [K_{xc}(z) + (\sigma + \sigma')J_{xc}(z) + \sigma\sigma' I_{xc}(z)].
\]

(2)

The φ<sub>σ</sub>(z) are self-consistent wave functions that diagonalise the effective one-dimensional LSDA Hamiltonian, after assuming translational invariance along the (x, y)-plane (area A). Indexes i, j, i', j' denote subbands and σ, σ' = 1(-1) for spin ↑(↓). V<sub>H</sub> in eq. (1) corresponds to the Hartree contribution with ε being the dielectric constant of GaAs, whereas V<sub>xc</sub> in eq. (2) corresponds to the exchange-correlation contribution with K<sub>xc</sub>, J<sub>xc</sub>, and I<sub>xc</sub> being derivatives of the exchange-correlation energy.
the charge and spin-density excitation $\hbar\omega_\mu$ can be written in the following compact way [4]:

$$\left(\hbar\omega_\mu\right)^2 - E_{i,i+1}^2 = 2 \delta n_{i,i+1} E_{i,i+1} \gamma_{i,i+1}^{x_{i,i+1}},$$

(3)

with $\delta n_{i,i+1} = n_i - n_{i+1}$ being the difference in the subband densities and $E_{i,i+1} = E_{i+1} - E_i$, where $E_i$ are the subband energies. In the paramagnetic ($P$) phase, the index $\mu = \text{CDE}$, SDE describes the uncoupled charge and spin density excitations. According to standard notations, $\gamma_{\text{CDE}}^{i,i+1} = (\alpha^{i,i+1}/\beta^{\text{CDE}})$, and $\gamma_{\text{SDE}}^{i,i+1} = -\beta^{\text{SDE}}$, provided the following identification is made: $\alpha = V_H$, $\beta_{\text{CDE}} = -(V_{xc}^{\uparrow\uparrow} + V_{xc}^{\downarrow\downarrow})/2$, and $\beta_{\text{SDE}} = -(V_{xc}^{\uparrow\downarrow} - V_{xc}^{\downarrow\uparrow})/2$. For the sake of simplicity, we have eliminated subband indexes, and we have used the symmetry $V_H = V_{xc}^{\uparrow\uparrow} = V_{xc}^{\downarrow\uparrow} = V_{xc}^{\downarrow\uparrow} = V_{xc}^{\uparrow\downarrow}$. $\Delta(\omega)$ is the contribution of the spin-density mode: it becomes softer, i.e., decreases in absolute value when the Fermi level is resonant with the second subband, recovering the normal behaviour at higher densities. From the study of the formation of a non-trivial magnetic order, intensively investigated in double-layer systems [17], a softening of the SDE can be considered as an indication of ferromagnetism in the second subband. In our system the softening is rather weak, probably due to the large contribution of electrons in the paramagnetic first subband. Comparing the three QW widths (fig. 2(a)) it can be seen that the SDE softening is enhanced for the narrowest QW. That speaks for a dependence of the possible ferromagnetic order on the strength of the wave function confinement. In fig. 2(b) we compare the experimental data (solid points) with the excitation energies yielded by TDLSDA calculations (empty points). The left (right) axis corresponds to the experimental (calculated) results. The underestimation of the softening resulting from the calculations can be attributed to shortcomings of the TDLSDA in the treatment of exchange and correlation but also to the fact that the calculations always yield a paramagnetic ground state for this high total density.

**Results.** – In fig. 2 we show an unexpected behaviour of the spin-density mode: it becomes softer, i.e., decreases in absolute value when the Fermi level is resonant with the second subband, recovering the normal behaviour at higher densities. From the study of the formation of a non-trivial magnetic order, intensively investigated in double-layer systems [17], a softening of the SDE can be considered as an indication of ferromagnetism in the second subband. In our system the softening is rather weak, probably due to the large contribution of electrons in the paramagnetic first subband. Comparing the three QW widths (fig. 2(a)) it can be seen that the SDE softening is enhanced for the narrowest QW. That speaks for a dependence of the possible ferromagnetic order on the strength of the wave function confinement. In fig. 2(b) we compare the experimental data (solid points) with the excitation energies yielded by TDLSDA calculations (empty points). The left (right) axis corresponds to the experimental (calculated) results. The underestimation of the softening resulting from the calculations can be attributed to shortcomings of the TDLSDA in the treatment of exchange and correlation but also to the fact that the calculations always yield a paramagnetic ground state for this high total density.

The key idea to exclude the influence of the highly filled (and paramagnetic) first subband, is to investigate higher intersubband excitations associated with the transition 1 → 2. These excitations occur between the sparsely populated second subband $E_1$ and the empty third subband $E_2$. Only electrons from the spin polarised phase in $E_1$ contribute to these higher excitations. In the series of spectra of fig. 1 the excitations SPE$_{12}$, SDE$_{12}$ and CDE$_{12}$ become apparent when the second subband is populated. In particular, the CDE$_{12}$ shows up exactly on top of the SPE$_{12}$ due to the collapse of the Hartree term $\alpha^{12}$ at very low densities [18]. In contrast, the SDE$_{12}$ is always shifted down from the SPE$_{12}$. From the measured energies of the excitations obtained with cross polarisation and using eq. (3) we determine the many-body correction energy $(2n_1(\gamma^{12})$. Multiplying by $n_1$ we avoid divergencies when the density goes to zero. The results are shown in fig. 3. The striking result of this work is the observation of an enhancement of the exchange-correlation contribution at low temperature when the system enters the region of the spin-instability. This enhancement leads to a pronounced softening of the collective mode...
associated with electronic transitions $1 \rightarrow 2$. The diluted electron gas of the second subband becomes thus unstable against spin-flip excitations, which trigger the transition into the ferromagnetic phase. By increasing the temperature a scaling-down of the exchange-correlation energy occurs, in accordance with the behaviour that one would expect for paramagnetic electron gases.

For a better understanding of the physical origin of this transition it is instructive to perform the corresponding calculation of the excitations. To avoid the effect of the high density in the first subband we constructed an auxiliary structure, using for its intersubband energy $E_{01}$ and density $n_0$, the experimental values $E_{12}$ and $n_1$, respectively. Doing so, the calculations can yield now a ferromagnetic ground state and the corresponding excitations.

Figure 4 compares the experimental $\gamma_{12}^{\text{exp}}$ values, as obtained from the excitations measured in cross polarisation at 2 K, with the calculated ones. TDLSDA predicts the occurrence of a ferromagnetic phase, characterised by $\gamma_M$ (open circles), for densities below $0.34 \times 10^{11}$ cm$^{-2}$ and a paramagnetic phase otherwise, characterised by the excitonic shifts $\beta_{\text{CDE}}$ and $\beta_{\text{SDE}}$ (triangles). Inspection of the different terms in $\gamma_M$ reveals that its strong increase at low densities is mainly due to the fact that the contribution from correlation has the opposite sign than the one to $\beta_{\text{SDE}}$ in the paramagnetic phase. Thus, correlation adds to the effect of exchange in the spin-polarised case, which favours ferromagnetism. In fact, the calculated values of $\beta_{\text{SDE}}$ and $\gamma_M$ at high and low densities, respectively, agree well with $\gamma_{12}^{\text{exp}}$. The range of stability of the ferromagnetic solution is also in nice agreement with the data of fig. 3, where the strong enhancement of exchange-correlation vertex corrections, observed at low temperatures, occurs at densities below $1 \times 10^{11}$ cm$^{-2}$.

In conclusion, we have obtained from light scattering experiments and local spin-density calculations a compelling evidence for the existence of a ferromagnetic ground state of two-dimensional electron gases in an effectively diluted regime.

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REFERENCES

[1] Bloch F., Z. Phys., 57 (1929) 545.
[2] Ceperley D. M., Nature, 397 (1999) 386.
[3] Reboredo F. A. and Proetto C. R., Phys. Rev. Lett., 79 (1997) 000463.
[4] Bolcatto P. G., Proetto C. R. and Reboredo F. A., Phys. Rev. B, 67 (2003) 073304.
[5] Jurin L. O. and Tamborenea P. I., Eur. Phys. J. B, 45 (2005) 9.
[6] Young D. P., Hall D., Torreli M. D., Fisk Z., Sarrao J. L., Thompson J. D., Ott H.-R., Oseroff S. B., Goodrich R. G. and Zysler R., Nature, 397 (1999) 412.
[7] Ghosh A., Ford C. J. B., Pepper M., Beere H. E. and Ritchie D. A., Phys. Rev. Lett., 92 (2004) 116601.
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[8] Goñi A. R., Giudici P., Reboredo F. A., Proetto C. R., Thomsen C., Eberl K. and Hauser M., Phys. Rev. B, 70 (2004) 195331.

[9] Abrahams E., Anderson P. W., Licciardello D. C. and Ramakrishnan T. V., Phys. Rev. Lett., 42 (1979) 673.

[10] Goñi A. R., Haboeck U., Thomsen C., Eberl K., Reboredo F. A., Proetto C. R. and Guinea F., Phys. Rev. B, 65 (2002) 121313(R).

[11] Reboredo F. A. and Proetto C. R., Phys. Rev. B, 67 (2003) 115325.

[12] Rigamonti S., Reboredo F. A. and Proetto C. R., Phys. Rev. B, 68 (2003) 235309.

[13] Pinczuk A., Schmitt-Rink S., Danan G., Valladares J. P., Pfeiffer L. N. and West K. W., Phys. Rev. Lett., 63 (1989) 1633.

[14] Gammon D., Shanabrook B. V., Ryan J. C. and Kitzer D. S., Phys. Rev. B, 41 (1990) 12311.

[15] Kohn W. and Vahistha P., Theory of the Inhomogeneous Electron Gas, edited by Ludqvist S. and March N. H. (Plenum, New York) 1983.

[16] Kohn W., Rev. Mod. Phys., 71 (1999) 1253.

[17] Bolcatto P. G. and Proetto C. R., Phys. Rev. Lett., 85 (2000) 1734.

[18] Ernst S., Goñi A. R., Syassen K. and Eberl K., Phys. Rev. Lett., 72 (1994) 4029.