Thermodynamic magnetization of two-dimensional electron gas measured over wide range of densities

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We report measurements of $\partial m/\partial n$ in Si MOSFET, where $m$ is the magnetization of the two-dimensional electron gas and $n$ is its density. We extended the density range of measurements from well in the metallic to deep in the insulating region. The paper discusses in detail the conditions under which this extension is justified, as well as the corrections one should make to extract $\partial m/\partial n$ properly. At low temperatures, $\partial m/\partial n$ was found to be strongly nonlinear already in weak magnetic fields, on a scale much smaller than the characteristic scales, expected for interacting two-dimensional electron gas. Surprisingly, this nonlinear behavior exists both in the dielectric, and in the metallic region. These observations, we believe, provide evidence for strong coupling of the itinerant and localized electrons in Si-MOSFET.

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Magnetic properties of strongly-interacting electron gas have long been a subject of intensive theoretical and experimental investigations. Coulomb interaction through the exchange term favors parallel spins, and therefore leads to ferromagnetism, whenever is strong enough. The strength of the interaction is determined by the ratio between the typical Coulomb and kinetic energies, and customarily characterized by the ratio between the typical inter-electron distance $\rho$, $\rho = 1/\sqrt{\pi n}$ in two dimensions, and the effective Bohr radius: $r_*=\rho/a_B$ \cite{1}. When the carrier density $n$ decreases, the relative effect of the interactions becomes stronger, contrary to the naive expectations. At sufficiently low density a clean system of itinerant electrons was predicted to become ferromagnetic, the phenomenon called Stoner instability \cite{2}; though in two dimensions it can occur only at zero temperature.

A real system of itinerant electrons is always disordered to some extent. Interactions, in the limiting case of strong disorder, favor antiparallel arrangements of the neighboring spins. The ground state of a pair of localized spins is a singlet, much like a Hydrogen molecule. A system of many localized spins preserves the tendency to antiferromagnetic order, see \cite{3} for review. Intensive investigation of the magnetic properties of the doped semiconductors, particularly phosphorus doped Si in 80s \cite{3}, lead to substantial understanding of the interplay between interactions and disorder. The observed divergency of the susceptibility with decreasing temperature at the low-density side of the metal-insulator transition was well understood; what remained unanswered, is the divergent susceptibility at the high-density side of the transition.

Interactions in 2D systems are more important than in 3D, which explains the wealth of experimental observations and difficulty in their explanation. Unfortunately, extremely small number of electrons hinders thermodynamic measurements of 2D systems. On the other hand, two-dimensional gated structures, particularly Si-MOSFETs, provide unique possibility of gradually changing the electron density, and, thus, interaction strength. Interest in the magnetic properties of 2DEG was sparked by the observation of strong suppression of conductivity in Si-MOSFETs by in-plane magnetic field \cite{4}. The strong magnetoresistance was interpreted in \cite{5, 6} as a quantum phase transition into a ferromagnetic state at the density of the metal-insulator transition, $n_c$. This interpretation was contested in \cite{7, 8} on the basis of Shubnikov-de Haas measurements, and in \cite{9} on the basis of thermodynamic magnetization measurements.

In the thermodynamic method, developed in \cite{10}, recharging current, generated between the 2DEG and the gate in response to modulation of external magnetic field $B$, is used to determine the derivative of the 2DEG chemical potential $\mu_{2D}$ with respect to the field. This derivative, by virtue of the Maxwell relation $\partial \mu/\partial B = -\partial m/\partial n$, can be translated into the derivative of the magnetic moment $m$ at a given field with respect to the density $n$. One can, in principle, integrate $-\partial m/\partial n$ over $n$ to get $m(n)$, assuming $m$ is known at a certain density. In Refs.\cite{9, 10} the integration constant was taken from transport measurements at some high density value.
It would be desirable to abandon this assumption by straightforward integration from $n = 0$, since at zero density the magnetic moment of an electron gas is zero by definition. Such approach, however, requires the measurements to be performed down to very low carrier densities, lower than were accessible earlier. In the present paper we extended the thermodynamic measurements down to densities as low as 0.3 $n_c$, deep in the insulating region. This extension enabled us to reveal strong unexpected nonlinearity of $\partial n/\partial V$ in weak magnetic fields, which, surprisingly, exists also in the metal phase.

Before presenting the experimental results, we first discuss in detail the way we extended the measurements, as well as the corrections which should be applied to properly interpret the data. Consider a system consisting of a 2DEG connected through an ohmic contact and a battery, which provides the gate voltage $V$, to the gate, as shown in Fig. 1. The free energy of the system can be written as follows:

$$f = f_G + f_{2D} - enV + \frac{e^2 n^2}{2\epsilon_0},$$

where $f_G$ and $f_{2D}$ are the free energies of the gate and the 2DEG, respectively, and the last term describes the electrostatic interaction between the 2DEG and the gate electrons. The capacitance $c_0$, defined by Eq. (1), differs from the geometric one by a factor of $\approx (1 + z_0/d_{\text{ox}})^{-1}$; here $d_{\text{ox}} \approx 190$ nm is the oxide thickness for the studied Si-MOSFETs, and $z_0 \approx 3.5$ nm is the average distance of the 2D electron layer from the Si/SiO$_2$ interface.

The minimum of $f$ determines the equilibrium density of the 2DEG. Differentiating Eq. (1) with respect to $n$ under the constrain of zero net charge of the system (the variation of the electron density at the gate $\delta n_g = -\delta n$) gives the familiar expression:

$$n = \frac{c_0}{e} \left( V - \frac{\mu_{2D} - \mu_G}{e} \right)$$

where $\mu_{2D}$ and $\mu_G$ are the chemical potentials of the electrons in the 2DEG and the gate respectively. We ignored $c_0$ dependence on the density; accounting for it leads to small corrections for both $n$, and $\tilde{c}$ defined below, of the order of $(\partial c_0/\partial n)(n/c_0)$.

A change of an external parameter shifts the equilibrium density $n$. We assume $\mu_G$, the chemical potential of a thin Al film, to be independent of $B$ and $n$. Consider first the density response on a variation of $V$, i.e. $dn/dV \equiv \tilde{c}/e$, which defines the gate-to-2DEG capacitance $\tilde{c}$:

$$\tilde{c} = c_0 \left( 1 + \frac{c_0}{e^2} \frac{\partial \mu_{2D}}{\partial n} \right)^{-1}$$

As seen from Eq. (3), $\tilde{c}$ is slightly renormalized, by a finite 2DEG compressibility.

Similarly, the density response to magnetic field variation is given by:

$$\frac{e^2}{\tilde{c}} \frac{dn}{dB} = -\frac{\partial \mu_{2D}}{\partial B} + \frac{e^2 n}{c_0^2} \frac{\partial c_0}{\partial B} \equiv -\frac{\partial \mu}{\partial B}.$$  

By measuring the recharging current in response to the gate voltage modulation one can get the capacitance $\tilde{c}$, and then use it to extract $\partial \mu/\partial B$. In Eq (4) we defined $\mu$, which contains an additional contribution proportional to $\partial c_0/\partial B$, neglected in [9] [10]. As we shall see below, it is indeed small for a 2DEG in Si-MOSFETs.

Since the thickness of 2DEG is finite, albeit small, an in-plane magnetic field couples also to the orbital motion, leading to two effects: (i) a diamagnetic shift of the spatial quantization levels [9] [12] [13]; (ii) a change in the average distance $z_0$ of the electrons from the Si-SiO$_2$ interface due to the asymmetry of the confining potential, see Fig. 1. The former effect causes a diamagnetic contribution to the chemical potential shift, $\delta \mu_d$, which should be subtracted, if one is interested in the spin contribution only: $\delta \mu_s = \delta \mu_{2D} - \delta \mu_d$. The later effect leads to a change of the capacitance $c_0(B)$, and results in the contribution $\delta \mu_c$, which should be subtracted from $\delta \mu$ in order to get $\delta \mu_{2D}$:  

$$\frac{\partial \mu_c}{\partial B} = -\frac{n e^2}{c_0^2} \frac{\partial c_0}{\partial B}.$$  

For a non-interacting 2D gas these finite thickness (FT) contributions can be found by solving numerically the Schrödinger equation in a triangular potential. We are interested in the energy of the lowest level $|0\rangle$ of the spatial quantization, since it is the only one populated at the experimentally relevant densities. We choose Landau gauge $\vec{A} = (0, B(z - z_0), 0)$, where $z_0 = (0|z|0)$.  

![Fig. 1. A sketch of the experimental setup, superimposed on the band diagram of a Si MOSFET sample; z-direction is perpendicular to the 2DEG plane](image)
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For density-dependent confining electric field $k$ components with different $B$ they are shown in Fig. 2. For $B > 0$ the diamagnetic contribution does not exceed 0.10 to average the result over the in-plane $k$ vectors of the 2DEG, and the capacitance one is negative (it is shown in Fig. 2 with negative sign).

Such a gauge preserves the minimum of the dispersion relation $\bar{c}_0(k_z, k_y)$ at $k_y = 0$. Care should be taken to average the result over the in-plane $k$ vectors of the populated states to get $\bar{c}_0 = \langle \bar{c}_0 \rangle_k$ and $\bar{z}_0 = \langle \bar{z}_0 \rangle_k$, since components with different $k_y$ are affected differently by the field. For density-dependent confining electric field $E$ we adopted the expression [1]

$$ E = \frac{4e\pi}{\varepsilon_{Si}} \left( n_D + \frac{11}{32} n \right), \quad \text{(6)} $$

where $e \cdot n_D$ is the depletion region charge, $n_D \approx 10^{11} \text{ cm}^{-2}$ for our samples. At low, compared to the Fermi energy, temperatures the diamagnetic contribution $\partial \mu_d/\partial B = \partial^2(\bar{n}\bar{c}_0)/\partial n \partial B$. The capacitance contribution, Eq. (4) can be expressed through $\bar{z}_0$ as:

$$ \frac{\partial \mu_d}{\partial B} = n e^2 \frac{\partial(1/\bar{c}_0)}{\partial B} = \frac{4\pi ne^2}{\varepsilon_{Si}} \frac{\partial \bar{z}_0 }{ \partial B}. \quad \text{(7)} $$

Both contributions, being the first derivatives of even functions, are linear in $B$ at weak magnetic fields; they are shown in Fig. 2. For $B > 0$ the diamagnetic contribution is positive, and the capacitance one is negative (it is shown in Fig. 2 with negative sign). The sum of the FT contributions is positive, therefore disregarding them would lead to underestimation of $\partial m_+/\partial n = -\partial \mu_d/\partial B$. For our system the resultant contribution does not exceed 0.05 $\mu_B$ at $B = 1$ T at low densities and drops with $n$; therefore it is small compared to the measured $\partial \mu_d/\partial B$, at least for low temperatures, see Fig. 3. The FT contributions should be more important for systems with lower effective mass in $z$-direction, such as GaAs based heterostructures, or in higher magnetic fields [9].

When the gate voltage or the magnetic field is modulated at a sufficiently small frequency $\omega$, so that the system stays in the thermodynamic equilibrium, the modulation leads to a recharging current with the amplitude

$$ (a) \quad I_V = i\omega C(\omega) \delta V \quad \text{and} \quad (b) \quad I_B = -\frac{i\omega C(\omega)}{e} \frac{\partial \mu_d}{\partial B} \delta B, \quad \text{(8)} $$

respectively, where the capacitance $C = \delta E$ is proportional to the sample area $S$; Eqs. (8) were used in [9, 10], in order to determine the capacitance [5,k], and then to extract $\partial \mu_d/\partial B$ [5,a]).

However, for low electron densities, recharging of the sample is hindered by the large contact and 2DEG resistances. When the resistance approaches $1/\omega C$, the capacitance becomes complex, and its magnitude drops. We show below that, under certain assumptions, Eqs. (8) can still be used for extracting $\partial \mu_d/\partial B$ using the complex capacitance, provided both $I_V$ and $I_B$ are measured at the same conditions: frequency, magnetic field and temperature.

Recharging of the capacitor, either by modulation of the gate voltage or of the magnetic field, leads to gradients of the potential and the carrier density over the 2DEG area, and the electrochemical potential $\mu_d$ becomes a function of coordinates. In addition, recharging current leads to a potential drop across the contact. The current density $\vec{j}$ is governed by the continuity equation:

$$ e \frac{d n}{dt} = -\nabla \vec{j} = \nabla \left( \sigma \nabla \frac{\mu_d}{e} \right), \quad \text{(9)} $$

where $\sigma$ is, in general, a coordinate-dependent conductivity of the 2DEG and the contact region.

Following [14] we assume that the system stays in quasi-thermodynamic equilibrium, and that the characteristic spatial scale of the electrochemical potential variations is large compared to the oxide thickness. Then, Eq. (2) with coordinate-dependent $\mu_d(r)$ is satisfied locally, the assumption called the “local capacitance approximation”. Under this assumption, according to Eqs. (3) and (4), both voltage and magnetic field modulation generate variation of the density, with the only difference being $e\delta V$ replaced with $-\partial \mu_d/\partial B \delta B$. These variations enter as the source term in Eq. (2), whose Green’s function, subject to the proper boundary conditions, determines the current through the contact. This means that Eqs. (5) hold under local capacitance approximation, with the same $C$ in Eqs. (5,k) and (5,)).

We present results obtained with the most extensively measured Si-MOSFET sample with the peak mobility of 3.4$\text{m}^2/\text{Vs}$ at 1.7K. This sample is similar to that used in Ref. [4]; the metal-insulator transition in it occurs at a critical density $n_c \approx 8.5 \cdot 10^{10} \text{cm}^{-2}$. The results obtained with many other samples of similar quality were similar. Most of the measurements

Fig. 2. Finite thickness contributions to $\partial \mu_d/\partial B$ at $B = 1$ T; dashed-dotted line is the diamagnetic contribution $\partial \mu_d/\partial B$, dotted line – $\partial \mu_c/\partial B$, solid line – the sum of the two. Dashed line is $\partial \bar{z}_0 / \partial B$ (right scale, note that $\bar{z}_0 \approx 3.5$ nm).
were done with in-plane magnetic field modulated at frequency 6.2 Hz with 40 mT amplitude. The results scaled linearly with modulation amplitude. Figure 3 shows $\partial\vec{\mu}/\partial B$ as a function of the magnetic field. It is antisymmetric with respect to magnetic field $\partial\vec{\mu}/\partial B$ is frequency-independent up to 30 Hz. The upper measurement frequency is limited by the EMF in wire loops, as well as by eddy current heating.

From now on let us discuss the results for $B \geq 0$. As seen in Fig. 3 at the lowest temperature of 1.7 K, $\partial\vec{\mu}/\partial B$ is strongly nonlinear. For densities deep in the insulating regime, e.g. at $n = 5 \cdot 10^{10}$ cm$^{-2}$, as the field increases, $\partial\vec{\mu}/\partial B$ sharply drops, then reaches a minimum, and saturates, or even increases at larger fields. The value of $\partial\vec{\mu}/\partial B = -\partial m/\partial n$ at the minimum is approximately $-0.7 \mu_B$, indicating almost full spin polarization; at even lower densities it reaches a value of almost $-\mu_B$. In the metallic regime, $\partial\vec{\mu}/\partial B$ changes sign. In Fig. 3 we plotted $\partial\vec{\mu}/\partial B$ for $n = 2 \cdot 10^{11}$ cm$^{-2}$, the density at which it is maximal. Although the saturation value at this density is much smaller, about 0.15$\mu_B$, the saturation field $B^*$ is similar to the one for $n = 5 \cdot 10^{10}$ cm$^{-2}$. Note that $B^*$ is small, $\approx 0.6$ T, well below the polarization field for a non-interacting system ($\approx 11$ T), and for the interacting one ($\approx 2.7$ T), both estimated for $n = 1 \cdot 10^{11}$ cm$^{-2}$ [8, 7]. Such small $B^*$ implies an existence of a relevant energy scale, much smaller than the bare, and even interaction renormalized Fermi energy.

The decrease of $|\partial\vec{\mu}/\partial B|$ at the lowest temperature above $B^*$ can be attributed to the finite thickness contributions: the slope of the solid line in Fig. 3 reasonably agrees with the predicted FT contributions for this density. As temperature grows, the nonlinearity smears out, and its onset shifts to higher magnetic fields [12].

To explain qualitatively the results, one needs to invoke, besides itinerant electrons, states, localized either within the potential well [15], or in its close vicinity. These states can be recharged at the frequency of the measurements, and contribute to $\partial\mu_{2D}/\partial B$, but would not contribute to Hall conductivity or to Shubnikov-de Haas oscillations. The fact that the zero density gate voltage $V_0$ (i.e., the so called “threshold voltage”), determined from extrapolation of the Hall or Shubnikov-de Haas measurements, increases with reduction of electron mobility [1] indicates the existence of such states.

On the other hand, the simplistic two-liquid model, in which the non-interacting localized states exist independently of the interacting 2DEG, suggested in [15], cannot explain such a low value of $B^*$, since the characteristic field, $k_B T/\mu_B$, for polarization of a single spin (or non-interacting non-degenerate gas) at 1.7 K is 4.2 T, much bigger than $B^*$. We therefore believe, that any explanation of the presented results must involve interactions. Note, that the underlying mechanism should be different from the interactions between localized spins, discussed in [3], since such interactions lead to antiferromagnetic coupling, and slower than $1/T$ divergence of the susceptibility, in contrast with our observations [12]. We infer that coupling mediated by itinerant electrons should play a role.

Besides localized states, one can think about interaction-enhanced FT contributions. Indeed, the estimations above ignored interactions. The interaction energy, however, is comparable to the energy difference between the levels of spatial quantization in the well; e.g., for $n = 10^{11}$ cm$^{-2}$ both are about 10 meV. It is, therefore, not a priori clear that the interactions can be ignored in calculations of $\Delta z_0(B)$. One can envision a scenario, in which spin polarization leads to a change in $z_0$. Note that $\Delta z_0 \approx 1 \mu$m would suffice to contribute one Bohr magneton to $\partial\vec{\mu}/\partial B$. It would be, however, quite a coincidence if such a mechanism conspires to contribute $\partial\vec{\mu}/\partial B \approx -\mu_B$ at the saturation.

In conclusion, we succeeded in measuring the magnetic field dependence of $\partial\mu_{2D}/\partial B$ over a density range much wider than was accessible earlier, from metallic to deep in the insulating region. The dependence of $\partial m/\partial n$ on $B$ is found to be strongly nonlinear at low temperatures, already in magnetic fields much smaller than the characteristic fields expected for the 2D elec-

**Fig. 3.** $\partial\vec{\mu}/\partial B$ at $n = 5 \cdot 10^{10}$ cm$^{-2}$, deep in the insulating regime, full symbols, left scale; the temperatures are: ■ - 1.7 K • - 3.4K ▲ - 4.6K ▼ - 6.8K. □ - $\partial\vec{\mu}/\partial B$ at $n = 2 \cdot 10^{11}$ cm$^{-2}$ and 1.7 K, deep in the metallic region, right scale. The slope of the dashed line gives the derivative of susceptibility $-\partial x/\partial n$, and the slope of the solid line – estimated FT contributions, see Fig. 2.
tron gas. These results point to a strong coupling between the itinerant and localized states in high mobility Si-MOSFETs.

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