Anomalous Spin Polarization of GaAs Two-Dimensional Hole Systems

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We report measurements and calculations of the spin-subband depopulation, induced by a parallel magnetic field, of dilute GaAs two-dimensional (2D) hole systems. The results reveal that the shape of the confining potential dramatically affects the values of in-plane magnetic field at which the upper spin subband is depopulated. Most surprisingly, unlike 2D electron systems, the carrier-carrier interaction in 2D hole systems does not significantly enhance the spin susceptibility. We interpret our findings using a multipole expansion of the spin density matrix, and suggest that the suppression of the enhancement is related to the holes’ band structure and effective spin $j = 3/2$.

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I. INTRODUCTION

It was first pointed out by Janak that for a two-dimensional electron system (2DES) in a static magnetic field the exchange interaction acts like an effective magnetic field (in addition to the applied field) so that the Zeeman energy splitting is enhanced.\textsuperscript{1} Recently, the Zeeman splitting and spin susceptibility of interacting 2D carrier systems have been a subject of renewed interest\textsuperscript{2,3,4,5,6,7,8,9} fueled by the promise of a paramagnetic to ferromagnetic ground state transition at very low densities,\textsuperscript{10,11} and the possibility that the spin polarization is related to the apparent metal-insulator transition in dilute 2D systems.\textsuperscript{12} Experiments have mostly focused on determining the spin susceptibility from magneto-transport\textsuperscript{2,3,4,5,6,7,8,9} and magnetization\textsuperscript{10} measurements. The results generally show that the spin susceptibility of 2DESs in different materials, e.g., Si,\textsuperscript{2,3,4,5,6,7} GaAs,\textsuperscript{8,9} and AlGaAs\textsuperscript{9} increases as the density is reduced, one report\textsuperscript{13} even suggesting a ferromagnetic instability at the lowest densities.

Lately, the spin polarization of GaAs 2D hole systems (2DHSs) has become the subject of intensive research\textsuperscript{11,12,13,14,15,16,17} because the holes have a larger effective mass (than electrons) so that they can be made effectively more dilute while maintaining high quality. Furthermore, the spin polarization of holes is important in the context of ferromagnetic semiconductors such as GaMnAs where it is known that the ferromagnetism is mediated by the itinerant valence band holes.\textsuperscript{18,19,20} We show here that the spin susceptibility of 2DHSs depends dramatically on the shape of the confining potential. Moreover, we find that, in contrast to their 2D electron counterparts, dilute 2DHSs exhibit no significant enhancement of the spin susceptibility as compared with calculations which neglect exchange-correlation.\textsuperscript{21} We will argue that this surprising behavior is related to the holes’ band structure and the fact they have effective spin $j = 3/2$ rather than $j = 1/2$ which is the case for electrons.

| sample | carriers | structure | substrate | $n$ ($10^{10}$ cm$^{-2}$) | $\mu$ (m$^2$/Vs) |
|--------|----------|-----------|-----------|----------------|-----------------|
| H      | holes    | heterojunction (001) |             | 5.3             | 30              |
| Q1     | holes    | 150 Å wide QW (001) |         | 4.8             | 11              |
| Q2     | holes    | 200 Å wide QW (113)A |     | 6.8             | 55*             |
| A      | electrons| heterojunction (001) |             | 3.0             | 48              |

$^*\mu$ for $J \parallel [33\hat{2}]$. For $J \parallel [\hat{T}\hat{1}\hat{0}]$ we have $\mu = 35$ m$^2$/Vs.

II. SAMPLE PARAMETERS AND EXPERIMENTAL DETAILS

Four samples from different wafers, including two GaAs/AlGaAs heterojunctions and two GaAs quantum wells (QWs) flanked by AlGaAs barriers, were investigated in this study (Table I). Depending on their substrate orientation and carrier type, our samples were either Be-doped (samples H, Q1) or Si-doped (Q2, A). All samples were fitted with metal front and back gates to control their density as well as the electric field perpendicular to the 2D systems. We made measurements in $^3$He or dilution refrigerators down to a temperature $T = 0.03$ K and in magnetic fields up to 25 T.

III. EXPERIMENTAL RESULTS FOR (001) 2D HOLES

In Fig. (Ia) we show the longitudinal resistivity $\rho_{xx}$ versus in-plane magnetic field $B_\parallel$ for samples H and Q1 both measured at a density of $n = 3.7 \times 10^{10}$ cm$^{-2}$. The data shows a positive magnetoresistance with a marked change in functional form above the magnetic field $B_d$ that reflects the complete depopulation of the minority spin subband.\textsuperscript{13,14} In Fig. (Ia) $B_d$ is marked by arrows. Remarkably, the field $B_d$ depends greatly on the shape of the confining potential. Indeed, we have $B_d \approx 10.6$ T for sample H and $B_d \approx 20.5$ T for sample Q1, even though the data were taken at the same density. In Fig. (Ic) we

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FIG. 1: (a) Longitudinal resistivity $\rho_{xx}$ versus in-plane magnetic field $B_\parallel$ measured at $T = 0.3$ K for 2D hole samples, H and Q1, at the same density $n = 3.7 \times 10^{10}$ cm$^{-2}$. The depopulation fields $B_d$ are marked by arrows. (b) Calculated density $n_\parallel$ in the minority spin subband of samples H and Q1 as a function of $B_\parallel$. (c) Measured (squares) and calculated (solid line) depopulation field $B_d$ versus change $\Delta \xi$ of the electric field in sample H for constant density $n = 4.5 \times 10^{10}$ cm$^{-2}$.

FIG. 2: Measured (symbols) and calculated (lines) depopulation field $B_d$ as a function of $n$ for samples (a) H and (b) Q2. In (a), for the solid line and squares (dashed line and circles) $n$ was varied via a back (front) gate. In (b) the different symbols refer to $B_\parallel \parallel [\overline{1}01]$ and $B_\parallel \parallel [\overline{3}3\overline{5}]$ as indicated. The upper horizontal axes show the confinement parameter $r_s$ for the corresponding $n$. In (a) we used $r_s(n)$ for the front gate.

show $B_d$ in sample H when the electric field $E$ across the junction is varied by means of front and back gates such that $n$ is kept constant at $4.5 \times 10^{10}$ cm$^{-2}$. The field $B_d$ increases significantly with increasing $E$. In Fig. 2(a) we show the measured $B_d$ versus $n$ for sample H. The values of $B_d$ depend rather sensitively on whether $n$ is changed by means of a front or back gate.

IV. CALCULATIONS AND DISCUSSIONS

In order to explain the experimental results of Figs. 1 and 2 we have performed parameter-free calculations in the multiband envelope-function and self-consistent Hartree approximations for the quasi-2D system. Figure 1(b) shows the calculated density $n_\parallel$ in the minority spin subband as a function of $B_\parallel$. The lines in Figs. 1(c) and 2 show the calculated $B_d$ for the corresponding experiments. The calculations reproduce the different behavior of samples H and Q1 in satisfactory agreement with experiment.

A. Confinement potential dependence of $B_d$

As we discuss in the next section, the close agreement between the experimental and calculated $B_d$ in Figs. 1 and 2(a) is very surprising because the calculations do not take exchange-correlation effects into account. Such effects are indeed dominant for 2D electron systems that are as dilute as the 2DHSs of Figs. 1 and 2(a). Before elaborating on this aspect of our results, however, we first discuss the remarkably strong dependence of $B_d$ on the confining potential. For an ideal, strictly 2D system with effective mass $m^*$ and effective $g$ factor $g^*$ we have $B_d \propto 1/(m^*g^*)$ independent of the shape of the confining potential. To understand the surprising results in Figs. 1 and 2 we will first concentrate on the Zeeman splitting which gives rise to the dominant contribution of the confinement dependence of $B_d$ in 2DHSs. Then we discuss the effect of $B_\parallel$ on the orbital motion.

Unlike electrons in the conduction band that have spin 1/2, holes in the uppermost valence band are characterized by an effective spin 3/2 (Ref. 25). Subband quantization in 2DHSs yields a quantization of angular momentum with $z$ component $m = \pm 3/2$ for the heavy holes (HHs) and $m = \pm 1/2$ for the light holes (LHs). In our samples, only the lowest HH subband is occupied. The quantization axis of angular momentum that is enforced by HH-LH splitting points perpendicular to the 2D plane. The Zeeman energy splitting due to $B_\parallel$ thus competes with the HH-LH splitting and it is well-known that the $B_\parallel$-linear Zeeman splitting of HH states is suppressed. The simple model of Ref. 31 yields $B_d \approx 250$ T for the systems in Fig. 1(a).] In the following we will discuss why the depopulation fields $B_d$ observed in real 2DHSs are much smaller than what these arguments suggest.

The dispersion of HH states is known to be highly nonparabolic as a consequence of HH-LH coupling. Therefore, the suppression of Zeeman splitting linear in $B_\parallel$ is merely the lowest-order effect in a Taylor expansion of the spin-split dispersion $E_s(k_\parallel, B_\parallel)$ of HH states as a function of the (canonical) wave vector $k_\parallel$, $B_\parallel$, and spin index $\sigma$. Mixed higher-order terms proportional to $B_\parallel$ and $k_\parallel$ give rise to an average Zeeman splitting of the occupied hole states which is approximately linear in $B_\parallel$. Thus we find that $B_d$ is generally much smaller than the value one would expect if the $B_\parallel$-linear Zeeman splitting were suppressed. This is also consistent with previous experimental data for 2DHSs that were interpreted ignoring completely the suppression of $B_\parallel$-linear Zeeman splitting in HH systems.
Now we can understand why the Zeeman energy splitting in 2DHSs depends sensitively on the shape of the confining potential. The mixed higher-order terms that are responsible for the Zeeman energy splitting $E_Z(B_{||})$ of HH systems compete with the HH-LH splitting. The latter depends sensitively on the shape of the confining potential so that we have here a tool to tune $E_Z(B_{||})$ of 2DHSs. In narrow quasi-2D HH systems we have a large HH-LH splitting so that the Zeeman energy splitting is reduced, giving rise to a large $B_d$. We get a large $E_Z(B_{||})$ (a small $B_d$) in wide systems. We can define $E_Z(B_d)$ as the energy difference between the Fermi energy and the subband edge at $B_d$. In the wide heterojunction of Fig. 1(b), the calculated $E_Z(B_d)$ is 0.44 meV, significantly larger than $E_Z(B_d) = 0.26$ meV in the narrower QW, despite the smaller value of $B_d$ in the heterojunction. Similarly, the increase of $B_d$ with increasing $\Delta E$ in Fig. 1(c) reflects the change of the HH-LH splitting in the system.

Next we discuss the effect of $B_{||}$ on the orbital motion. In general, the mass of the particles in quasi-2D systems increases as a function of $B_{||}$ which reflects the fact that, ultimately, for large $B_{||}$ resulting in a magnetic length comparable to the width of the quasi-2D system the particle states become dispersionless Landau levels. Obviously, this effect depends on the thickness of the quasi-2D system and it has been shown that $B_d$ in wide quasi-2D electron systems is much smaller than $B_d$ in narrow 2DESs. We will argue next that the mass enhancement does not explain, however, the results in Figs. 1 and 2(a).

Our numerical calculations show, in agreement with the 2DESs’ results, that the mass enhancement at small $B_{||}$ is smaller in the QW than in the heterojunction. However, $m^*_{\parallel}$ in 2DHSs increases highly nonlinearly as a function of $B_{||}$ which is particularly important for the QW with the larger $B_d$. Thus we find that at $B_d$ the mass enhancement in the narrower 2DHS of the QW is larger than in the wide 2DHS of the heterojunction. We note that at $B_d$ the mean kinetic energy equals approximately half the Zeeman energy splitting $E_Z(B_d)$ so that for Fig. 1(b) the mass enhancement can be inferred from the $E_Z$ values quoted above [see also Eq. 1 below].

The anomalous enhancement of $m^*$ at $B_d$ with decreasing width of the quasi-2D HH system depends sensitively on the system parameters such as the density and the shape of the confining potential. For the parameters in Fig. 1(c), $m^*$ at $B_d$ is approximately independent of $\Delta E$ (despite the significant change of $B_d$), i.e., the increase of $B_d$ with $\Delta E$ is essentially only due to the decrease of the Zeeman splitting discussed above. For about twice the largest field $\Delta E$ we could reach experimentally one enters the regime when $m^*$ at $B_d$ starts to increase with $\Delta E$.

A most remarkable aspect of the results in Figs. 1 and 2(a) is the reasonable quantitative agreement between the experimental data and the calculations. This is particularly puzzling because many-particle effects beyond the Hartree approximation (i.e., exchange-correlation effects) were not taken into account. This is in sharp contrast to the case of wide 2DESs for which it is known that exchange-correlation significantly increases the spin susceptibility when $n$ is reduced. To quantify this point we show in Fig. 3 the ratio $B^{0}_d/B^{\exp}_d$ of the depopulation field $B^{0}_d$, calculated neglecting exchange-correlation, to the experimentally measured field $B^{\exp}_d$ for a 2DES (squares) and the 2DHS (circles) in sample H. The ratio thus reflects the enhancement of the spin susceptibility at $B_d$ due to exchange-correlation. Our results are plotted as a function of the dimensionless density parameter $r_s$ defined as the average interparticle spacing measured in units of the effective Bohr radius $a_0^{\parallel}$, $r_s \equiv 1/(a_0^{\parallel}\sqrt{\pi n})$.

For the 2DES in Fig. 3 the ratio $B^{0}_d/B^{\exp}_d$ is between 2 and 4 and, as expected, it increases with $r_s$. We also remark that for electrons the experimentally observed reduction of $B_d$ is in reasonable quantitative agreement with numerical calculations that take exchange-correlation into account. To illustrate this point, the solid line in Fig. 3 shows the ratio $B^{0}_d/B^{\exp}_d$ where $B^{\exp}_d$ was calculated in the framework of spin-density-functional theory using a parameterization of the polarization-dependent exchange-correlation potential that was recently obtained by means of quantum Monte Carlo calculations. For the 2DHS, on the other hand, the expected enhancement of the spin susceptibility and $B^{0}_d/B^{\exp}_d$ ratio is conspicuously absent in Fig. 3. Note that, because of their larger effective mass compared to GaAs electrons ($m^* \simeq 0.25$ compared to $m^* = 0.067$: here $m^*$ is given in units of the free-electron mass),
2DHSs have significantly larger \( r_s \) and are thus effectively much more dilute. Nonetheless, the ratio \( B_0^0/B_0^\exp \) remains close to unity up to the largest values of \( r_s \) where a greater than ten-fold enhancement is expected.

Before discussing possible reasons for this anomalous behavior of 2DHSs, we make remarks regarding the effective mass \( m^* \) which enters \( a_0 \) and thus determines \( r_s \). For holes, \( m^* \) is not uniquely defined. As discussed above, the HH dispersions are typically nonparabolic, meaning that \( m^* \) depends on energy and therefore on \( n \) and the confinement potential. Moreover, the HH systems have a large Rashba and Dresselhaus spin splitting at \( B = 0 \) (Ref. 28), leading to two energy versus wavevector (\( k_B \)) dispersions with different curvatures and effective masses \( m^*_n \) and \( m^*_\pm \). Commonly, values of \( m^* \) between about 0.2 and 0.4 are used for holes in GaAs. 11,14,15,25,34 Here we adopt a simple definition for an average effective mass \( \langle m^* \rangle \):

\[
\langle m^* \rangle = \frac{\hbar^2 \pi n}{2\langle E_k \rangle},
\]

where \( \langle E_k \rangle \) is the mean kinetic energy per particle. Figure 2(a) shows the calculated density parameter \( r_s \) in sample H, when \( n \) is changed by means of a front or back gate. Note that for a single, parabolic dispersion with an effective mass \( m^* \), the mass \( \langle m^* \rangle \) as defined in Eq. (1) properly reduces to \( m^* \) and is independent of \( n \). For the 2DHS, on the other hand, \( \langle m^* \rangle \) in general depends sensitively not only on \( n \) but also on the system’s parameters such as the thickness of the 2DHS and on the applied electric and magnetic fields, as discussed above. If we take into account Rashba and Dresselhaus spin splitting then we get, similar to Eq. (1), effective masses \( \langle m^* \rangle \) for each spin subband. To illustrate this effect, we show \( \langle m^* \rangle \) for sample H in Fig. 2(b). We emphasize that the main conclusion of our work, namely the lack of enhancement of the spin susceptibility with increasing diluteness, is not affected by the specific values of \( m^* \) used to define \( r_s \); it is clear in Fig. 2(b) that if \( r_s \) were changed by a factor of 2 or 3, there would still exist a large discrepancy between the experimental hole data and the expected enhancement.

Why do dilute 2DHSs not show a significant enhancement of the spin susceptibility? Using a recently developed multipole expansion of the spin density matrix, we argue in the remainder of this paper that the \( j = 3/2 \) hole spin is the likely culprit. For 2DESs with spin 1/2 it is well-known that the mean Coulomb energy \( \langle E_c \rangle \) per particle can be completely characterized using \( n \) and (the magnitude of) the spin polarization \( \zeta \) as independent parameters. This is because the \( 2 \times 2 \) spin density matrix of spin 1/2 systems can be decomposed into four independent terms: \( n \) (a monopole) and the three components of the spin polarization vector \( \zeta \) (a dipole). In the Hartree-Fock (HF) approximation, the direct part of the Coulomb energy \( \langle E_c^{HF} \rangle \) cancels the potential of the positive background so that only the exchange term \( \langle E_x \rangle \) remains.

\[
\langle E_c \rangle = \langle E_c \rangle = \frac{2e^2}{3\pi}\sqrt{\frac{2\pi n}{3}} \left[ (1 + \zeta)^{3/2} + (1 - \zeta)^{3/2} \right].
\]

The Coulomb energy \( \langle E_c^{HF} \rangle \) is thus proportional to \( \sqrt{n} \). Higher order terms in a series expansion for \( \langle E_c \rangle \) of 2DESs were calculated in Ref. 27, \( \langle E_c \rangle \) was calculated numerically in, e.g., Refs. 28 and 29. For sample H, the normalized dipole and octupole at \( B_d \) are shown in Figs. 4(c) and (d) (Ref. 35). Unlike 2DESs, the dipole at \( B_d \) is much smaller than unity, i.e., despite the fact that only one spin subband is occupied at \( B_d \), the system is only weakly spin polarized. This result is an immediate consequence of the suppression of the \( B_1 \)-linear Zeeman splitting discussed above. The octupole can be interpreted as a new “spin degree of freedom” of spin 3/2 hole systems at \( B > 0 \) (Ref. 16). For sample H, the normalized dipole and octupole at \( B_d \) are shown in Figs. 4(c) and (d) (Ref. 35). Unlike 2DESs, the dipole at \( B_d \) is much smaller than unity, i.e., despite the fact that only one spin subband is occupied at \( B_d \), the system is only weakly spin polarized. This result is an immediate consequence of the suppression of the \( B_1 \)-linear Zeeman splitting discussed above. The octupole can be interpreted as a new “spin degree of freedom” of spin 3/2 hole systems at \( B > 0 \) which does not exist for the more familiar case of spin 1/2 electron systems. When the spin polarization is suppressed for an in-plane magnetic field, the 2D HH systems acquire instead a large octupole moment as visible in Fig. 4(d). The quadrupole is always close to unity because the HH-LH mixing is small in the systems considered here. Therefore, the
quadrupole is not shown in Fig. 4. By definition, these four multipoles provide a set of independent parameters that can be used to parameterize the Coulomb energy \( \langle E_c \rangle \) of spin 3/2 systems, similar to \( \langle E_c \rangle (n, \zeta) \) in spin 1/2 systems.\(^{29}\) However, the series expansion is presently not known and its calculation represents a formidable task. Our study indicates that the series expansion of \( \langle E_c \rangle \) of spin 3/2 2DHs is qualitatively different from \( \langle E_c \rangle (n, \zeta) \) of spin 1/2 2DESs.

The HF exchange energy \( \langle E_x \rangle \) of 2D HH systems at \( B = 0 \) is the same as \( \langle E_x \rangle \) of spin 1/2 2DESs because Eq. (2) requires only that the eigenstates of the two spin subbands for the same \( k_{\parallel} \) are orthogonal.\(^{24}\) For a HH system, the main effect of a perpendicular magnetic field \( B_{\perp} \) is a spin polarization (a dipole), whereas an in-plane field \( B_{\parallel} \) usually gives rise to an octupole moment.\(^{24-26}\) The spin density matrices of 2D HH systems at \( B_{\perp} > 0 \) and \( B_{\parallel} > 0 \) are thus qualitatively different. However, the HF exchange energy does not distinguish between these cases and always leads to the same enhancement of the exchange energy as in 2DESs. We note that different results can be obtained for \( \langle E_x \rangle \) when HH-LH mixing is significant.\(^{24}\) Also, different results are obtained for \( \langle E_x \rangle \) in higher-order perturbation theory when the more complicated energy dispersion must be taken into account. These are the reasons why the well-established results for exchange-correlation in dilute spin 1/2 2DESs cannot easily be transferred to spin 3/2 2DHs.

V. RESULTS FOR (113) 2D HIGHER-ORDER MULTIPLE-LEVELS

We extend our investigation by comparing the results for sample H with the data for Q2, a QW grown on a (113)A GaAs substrate. Figure 2(b) shows \( B_d \) versus \( n \) for Q2. The field \( B_d \) strongly depends on whether \( B_{\parallel} \) is applied in the in-plane crystallographic directions \([1\overline{1}0] \) or \([3\overline{2}2] \).\(^{11}\) The right column of Fig. 2 shows \( n_s \), \( \langle n_s^+ \rangle \), the dipole and the octupole moments calculated for Q2. For this sample, when \( B_{\parallel} \) is applied parallel to \([3\overline{2}2] \), the measured \( B_d \) is well below the calculated value. It is remarkable that for this particular geometry, the octupole remains small, but the 2DHs develops a large dipole moment [Figs. 2(c) and (d’)], similar to 2DESs in a \( B_{\parallel} \).\(^{12}\) This observation suggests that the spin susceptibility is enhanced by many-particle effects only when the magnetic field gives rise to a spin polarization. On the other hand, Figs. 2 and 4 suggest that there is no significant enhancement in \( j = 3/2 \) 2DHSs with a large octupole but a small dipole moment.

VI. SUMMARY AND CONCLUSIONS

We have shown that the spin susceptibility of dilute GaAs 2DHs in an in-plane magnetic field \( B_{\parallel} \) depends sensitively on the shape of the confining potential. Most remarkably, the spin susceptibility is not significantly enhanced as compared with calculations which neglect the carrier-carrier interaction. This is in sharp contrast to dilute electron systems for which it is known that many-body effects greatly enhance the spin susceptibility. Using a multipole expansion of the spin density matrix we have argued that the suppression of the enhancement is related to the holes’ band structure and effective spin \( j = 3/2 \).

Our findings have important implications for the quantum phase diagram of dilute 2DHs. In dilute electron systems, the exchange-correlation enhancement of the spin susceptibility can be considered a precursor for the ferromagnetic liquid which is expected to be the ground state of ultra-low density 2DESs with \( r_s \gtrsim 26 \).\(^{29}\) The extra multipoles of 2DHs provide new possibilities for the ground state of hole systems to respond to external perturbations such as a magnetic field thus leading to a richer phase diagram than in spin 1/2 electron systems.\(^{26-28}\) However, our results suggest that a ferromagnetic phase (i.e., a fully spin-polarized phase with a maximum dipole moment) is often not favored in dilute 2DHs. This could also have important implications for ferromagnetic semiconductors such as GaMnAs where it is known that the ferromagnetism is mediated by the itinerant spin 3/2 holes in the valence band.\(^{29,30}\) In itinerant ferromagnets it is the polarization-dependent competition between the Coulomb energy and the kinetic energy of the interacting carriers which controls the ferromagnetic transition.

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In Fermi liquid theory, the enhancement of the spin susceptibility is proportional to the product $m^*_{QP}g^*_{QP}$, where $m^*_{QP}$ is the effective mass and $g^*_{QP}$ is the effective $g$ factor of the quasiparticles at the Fermi energy. Recently, several authors studied the many-body enhancement of $m^*_{QP}$ in electron systems, see, e.g., Y.-W. Tan et al., Phys. Rev. Lett. 94, 016405 (2005) and references therein. We note that these findings cannot easily be compared with hole systems because of the highly nonparabolic energy dispersion of holes. This is one of the reasons why we focus instead on the spin susceptibility when discussing many-body effects.

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In the calculations, front and back gate are modeled by different boundary conditions for the Hartree potential.

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