

## Exchange instabilities in semiconductor double-quantum-well systems

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We consider various exchange-driven electronic instabilities in semiconductor double-layer systems in the absence of any external magnetic field. We establish that there is no exchange-driven bilayer-to-monolayer charge-transfer instability in the double-layer systems. We show that, within the unrestricted Hartree-Fock approximation, the low-density stable phase (even in the absence of any interlayer tunneling) is a quantum “pseudospin-rotated” spontaneous interlayer phase-coherent spin-polarized symmetric state rather than the classical Ising-like charge-transfer phase. The  $U(1)$  symmetry of the double-quantum-well system is broken spontaneously at this low-density quantum phase transition, and the layer density develops quantum fluctuations even in the absence of any interlayer tunneling. The phase diagram for the double-quantum-well system is calculated in the carrier density-layer separation space, and the possibility of experimentally observing various quantum phases is discussed. The situation in the presence of an external electric field is investigated in some detail using the spin-polarized-local-density-approximation-based self-consistent technique and good agreement with existing experimental results is obtained. [S0163-1829(97)06708-8]

### I. INTRODUCTION

Exchange-driven instabilities in an electron gas have been a subject of long-standing interest<sup>1</sup> in condensed matter physics dating back to 1929, when Bloch first pointed out<sup>2</sup> that a low-density free electron gas may undergo a spontaneous spin polarization transition to a ferromagnetic state by virtue of the dominance of exchange energy over kinetic energy at low enough electron density. A number of possible exchange instabilities has been proposed and extensively studied theoretically<sup>1</sup> in three-dimensional electron systems including ferromagnetism, antiferromagnetism, and various spin and/or charge texture phases. It is, however, unclear whether any such exchange instability has ever been experimentally observed in a three-dimensional free-electron-like system. One problem is that the available three-dimensional free-electron systems, namely alkali metals, have reasonably high effective electron densities, making the normal paramagnetic ground state energetically stable and exchange instabilities unlikely.<sup>1</sup> Recent interest in this subject has focused on the possibility of exchange instabilities in two-dimensional electron systems as occurring in artificially structured semiconductor quantum wells, heterostructures, and superlattices. These two-dimensional electron systems, particularly the ones existing in modulation-doped GaAs- $\text{Al}_x\text{Ga}_{1-x}\text{As}$  nanostructures, offer several advantages over three-dimensional electron systems (e.g., metals, doped bulk semiconductors) in terms of a systematic study of exchange-correlation effects. First, lower dimensionality typically enhances interaction effects, making exchange instabilities more likely in two-dimensional electron systems. Second, the electron density can be varied over (almost) two orders of magnitude in modulation-doped two-dimensional systems (either by varying the modulation doping level and/or by using suitable gates), thereby enabling one to tune the relative magnitude of exchange-correlation effects. Third, these artificially structured two-dimensional systems can be made ultrapure (substantially reducing disorder ef-

fects) because the ionized dopants are spatially separated from the electron layer. Fourth, artificial structuring enables the introduction of additional degrees of freedom into the problem, e.g., separation between the layers in a bilayer system, which are not available in purely two- or three-dimensional electron systems, thus allowing the possibility of further tuning interaction effects. Finally, and perhaps most importantly, the application of a strong external magnetic field perpendicular to the two-dimensional layer quenches the kinetic energy of the system as the two-dimensional electron gas gets quantized into Landau levels, thereby increasing the importance of electron-electron interaction effects. Because of these reasons, as well as the obvious reason of substantial experimental and technological relevance, there has been a great deal of recent interest in the possibility of interaction (i.e., exchange-correlation) induced exchange instabilities in two-dimensional systems. While much of this recent activity<sup>3-10</sup> focuses on the situation in the presence of an external magnetic field, there has also been considerable interest<sup>10-17</sup> in the possibility of exchange instabilities in two-dimensional electron gases in the absence of any external magnetic fields. In this paper we theoretically investigate a specific zero magnetic field exchange instability, namely a charge-transfer instability, which has been predicted to occur in semiconductor double-quantum-well systems<sup>10,11,15-17</sup> under suitable conditions.

The basic issue we study is quite simple. Consider a semiconductor double-quantum-well structure (e.g.,  $\text{Al}_x\text{Ga}_{1-x}\text{As-GaAs-Al}_y\text{Ga}_{1-y}\text{As-GaAs-Al}_x\text{Ga}_{1-x}\text{As}$  system) at zero temperature, which has been modulation doped to produce a bilayer two-dimensional electron system (in the  $x$ - $y$  plane) with a layer separation  $d$  (in the  $z$  direction) and a *total* two-dimensional electron density  $2n$  (per unit area). Simple electrostatic considerations imply that the equilibrium situation, which minimizes the Coulomb energy, is a classically symmetric situation with each quantum well equally populated with an electron density  $n$ . (Quantum) kinetic energy is also minimized by having equal populations

of both layers, as this leads to a lower Fermi energy. Thus the naive expectation (which, as we shall prove in this paper, turns out to be correct in this case) is that the double-quantum-well system prefers a bilayer electron gas with each layer equally populated with electrons. It has, however, been pointed out<sup>10,11</sup> that this simple picture may break down at low density and small interlayer separation where there could be a zero-temperature (quantum) phase transition from a bilayer to a monolayer system driven entirely by exchange-correlation effects. This low-density bilayer-to-monolayer phase transition<sup>11</sup> is, in fact, an exchange instability where at some low values of  $n$  there is predicted to be a spontaneous charge transfer from one layer to another, resulting in a symmetry-broken monolayer phase where, instead of a bilayer electron system with each layer having an electron density of  $n$ , all the electrons reside in one layer with an electron density of  $2n$ . This transition is similar to the exchange-driven ferromagnetic spin polarization transition. This conclusion on the existence of a charge-transfer instability in a double-quantum-well system was reached in Ref. 11 by considering the competition among the kinetic energy, the Coulomb (Hartree) charging energy, and the exchange energy. Our goal is to investigate the problem in the unrestricted Hartree-Fock approximation (HFA) by treating the layer index as a fully quantum-mechanical variable. We conclude that there can be no exchange-driven charge-transfer instability in a semiconductor quantum-well system under any conditions. The suggested charge-transfer instability is a feature of the restricted HFA where the layer index is treated as a classical Ising-like variable. In the more general unrestricted HFA, there is an exchange-driven instability towards a transition to a low-density symmetric phase rather than the monolayer phase.

A related issue we investigate connects with the recent experimental search<sup>15-17</sup> to observe the predicted charge-transfer instability with some of the papers<sup>15</sup> reporting experimental support for an abrupt double-to-single-layer transition in a double-quantum-well structure. These experimental studies involve measurements of layer electron densities [via low field Shubnikov-de Haas (SdH) oscillations] in a double-quantum-well system *under the application of an external electric field*. The applied electric field explicitly breaks the layer symmetry in the problem, and the observed nonlinearity<sup>15-17</sup> in the layer depopulation is a direct manifestation of the so-called exchange-correlation induced “negative compressibility” effect.<sup>18</sup> We study the layer or subband electron densities in the GaAs double-quantum-well structures in the presence of an applied electric field within the self-consistent spin polarized local-density approximation, obtaining excellent agreement with the existing experimental measurements.<sup>15-17</sup> The same self-consistent approximation is used to calculate the phase diagram of the double-quantum-well system in the absence of any external electric field and no stable monolayer electronic phase is found.

The rest of this paper is organized as follows. In Sec. II we investigate the phase diagram of a double-quantum-well structure in the electron density ( $n$ )-layer separation ( $d$ ) space within the restricted HFA including effects of electron spin. In Sec. III we allow for the possibility that the layer electron density is not required to be a good quantum num-

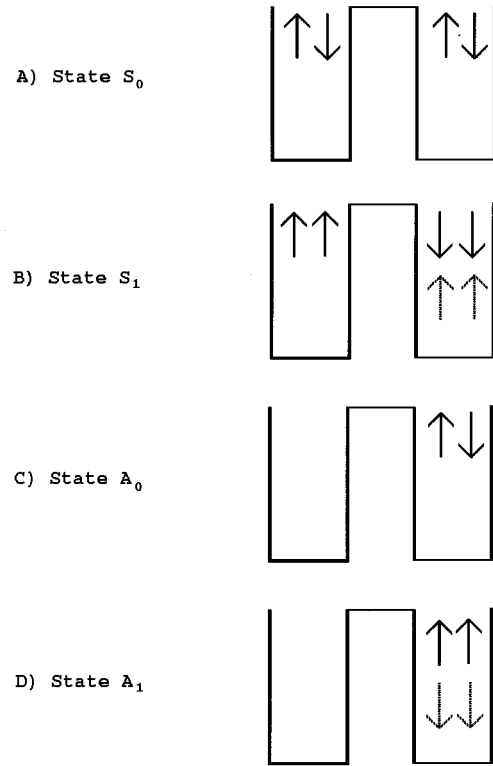


FIG. 1. The definitions of the four phases,  $S_0$ ,  $S_1$ ,  $A_0$ , and  $A_1$  used in the mean-field phase diagrams.

ber *even in the absence of interlayer tunneling* and explicitly include quantum fluctuations in the layer density by considering symmetric quantum states which are linear superpositions of electron states confined to different layers. Such a “pseudospin-rotated” quantum state, which involves no charge transfer, is shown to always have a lower energy than the monolayer charge-transfer phase, establishing unambiguously that the monolayer phase is *not* energetically stable in the HFA. In Sec. IV we consider the recent “charge-transfer” experiments in double-quantum-well systems in the presence of external electric fields, obtaining quantitative agreement between measured electron densities and self-consistent spin polarized local-density calculations. We conclude with a discussion in Sec. V.

## II. RESTRICTED HARTREE-FOCK APPROXIMATION

Consider a double-quantum-well system where each electron is in spin up or down and in layer left (or layer 1) or right (or layer 2) states (Fig. 1). In this system there are four possible (completely polarized or unpolarized) phases which are denoted  $S_0$  (equal population of both layer and spin components: the normal bilayer paramagnetic phase),  $S_1$  (equal population of both layers, but the electrons are spin polarized in each layer: the bilayer ferromagnetic phase),  $A_0$  (equal population of each spin component, but all the electrons are in a single layer: the monolayer paramagnetic phase), and  $A_1$  (the electrons are spin polarized and reside only in one layer: the monolayer ferromagnetic phase). Earlier work<sup>11</sup> did not explicitly consider the possibility of an exchange-driven spin polarization transition (considering only paramagnetic phases with equal populations of both up and down

spins) and therefore included only the possibility of  $S_0$  and  $A_0$  phases in their restricted HFA of double-quantum-well charge-transfer instability. The fundamental principle underlying the exchange instability is that exchange interaction prefers a spatially antisymmetric wave function which, by keeping the electrons away from each other, optimizes the interaction energy. This can be accomplished equally effectively by having a symmetric spin state (i.e., a spin polarized ferromagnetic state) and/or by having a symmetric layer state (i.e., a monolayer state), which will necessarily imply that the spatial part of the wave function is antisymmetric. Thus, exchange should lead to a spin polarization ferromagnetic transition in each layer as much as the bilayer-to-monolayer transition. In fact, the exchange-driven intralayer ferromagnetic transition ( $S_0 \rightarrow S_1$ ) is more likely than the bilayer-to-monolayer transition ( $S_0 \rightarrow A_0$ ) because there is no Coulomb Hartree energy to overcome in the spin polarization transition. With this introduction to the possible spin and layer phases of the double-quantum-well system, we discuss the HFA to the ground-state energy including only kinetic, exchange, and Hartree energy contributions. Following Refs. 10 and 11, we model each electron layer as a two-dimensional sheet of zero thickness, which then allows for a simple analytic calculation of the ground-state HFA energy per unit area,  $E^{\text{HF}}(n, d)$ , as a function of the layer separation  $d$  and the electron density  $n$ .

For a two-layer system with  $n_i$  and  $m_i$  being, respectively, the electron density and the spin polarization index of layer  $i$  ( $i=1$  or  $2$ ), the total energy per unit area within the HFA is [here  $m=(n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow)$  is the spin polarization in a particular layer, with  $n_{\uparrow(\downarrow)}$  being the number density of spin up (down) electrons in the layer]

$$E^{\text{HF}} = \frac{e^2 n_1}{2a^*} \left( \frac{1+m_1^2}{r_{s1}^2} - \frac{4\sqrt{2}}{3\pi r_{s1}} [(1+m_1)^{3/2} + (1-m_1)^{3/2}] \right) + \frac{e^2 n_2}{2a^*} \left( \frac{1+m_2^2}{r_{s2}^2} - \frac{4\sqrt{2}}{3\pi r_{s2}} [(1+m_2)^{3/2} + (1-m_2)^{3/2}] \right) + \frac{2\pi e^2 d}{\kappa_{\text{barrier}}} \left( \frac{n_2 - n_1}{2} \right)^2, \quad (1)$$

where  $r_{si} = 1/(a^* \sqrt{\pi n_i})$ , with  $a^* = \kappa \hbar^2 / m^* e^2$ , where  $\kappa = (\kappa_{\text{well}} + \kappa_{\text{barrier}})/2$  is the lattice dielectric constant, as the effective Bohr radius for the double-quantum-well system. Note that, in general,  $0 \leq n_i \leq 2n$  with the constraint  $n_1 + n_2 = 2n$ , and  $0 \leq |m_i| \leq 1$ . The various contributions in Eq. (1) for the HFA energy are the kinetic energy (the two terms involving  $r_s^{-2}$ ), the exchange energy (the two terms involving  $r_s^{-1}$ ), and the electrostatic Hartree Coulomb energy associated with charge transfer (the last term).

For the state  $S_0$ ,  $n_1 = n_2 = n$  and  $m_1 = m_2 = 0$ ,

$$E^{\text{HF}} = 2 \left( \frac{1}{r_s^2} - \frac{8\sqrt{2}}{3\pi r_s} \right) \frac{e^2 n}{2a^*}. \quad (2)$$

For the state  $S_1$ ,  $n_1 = n_2 = n$  and  $m_1 = -m_2 = 1$ ,

$$E^{\text{HF}} = 2 \left( \frac{2}{r_s^2} - \frac{16}{3\pi r_s} \right) \frac{e^2 n}{2a^*}. \quad (3)$$

For the state  $A_0$ ,  $n_1 = 0$ ,  $n_2 = 2n$ , and  $m_1 = m_2 = 0$ ,

$$E^{\text{HF}} = \left[ 2 \left( \frac{2}{r_s^2} - \frac{16}{3\pi r_s} \right) + \frac{4d}{a} \frac{1}{r_s^2} \right] \frac{e^2 n}{2a^*}. \quad (4)$$

For the state  $A_1$ ,  $n_1 = 0$ ,  $n_2 = 2n$ ,  $m_1 = 0$ , and  $m_2 = 1$ ,

$$E^{\text{HF}} = \left[ 2 \left( \frac{4}{r_s^2} - \frac{16\sqrt{2}}{3\pi r_s} \right) + \frac{4d}{a} \frac{1}{r_s^2} \right] \frac{e^2 n}{2a^*}. \quad (5)$$

In Eqs. (4) and (5),  $a = \kappa_{\text{barrier}} a^* / \kappa$  is different from  $a^*$  because  $\kappa_{\text{well}} \equiv \kappa_{\text{GaAs}} \neq \kappa_{\text{barrier}} \equiv \kappa_{\text{Al}_x\text{Ga}_{1-x}\text{As}}$  in the double-quantum-well system. (The quantitative correction arising from this difference is very small since  $a^* = 98.3 \text{ \AA}$  and  $a = 95.6 \text{ \AA}$ .)

Before presenting our results, we make some brief remarks about Eqs. (1)–(5). First, we note that there is some arbitrariness in our definitions of the symmetry-broken phases  $S_1$ ,  $A_0$ , and  $A_1$ . In particular, each spin polarized phase ( $S_1, A_1$ ) is characterized by  $|m| = 1$ , and therefore we could choose, for example, for  $S_1$ ,  $m_1 = m_2 = 1$ , and for  $A_1$ ,  $m_1 = 0$ ,  $m_2 = -1$ . It is obvious that this arbitrariness does not affect energetics and the calculated phase diagram, and is just the usual arbitrariness of the order parameter in the broken symmetry phase. Second, we point out that the symmetry broken phases  $S_1$ ,  $A_0$ , and  $A_1$  are completely spin and/or layer polarized phases where the symmetry-broken order parameters ( $m_i, n_i$ ) take on their maximum (in magnitude) values allowed (i.e.,  $|m_i| = 1$ ,  $|n_i| = 2n$ ). In general, partial spin and/or layer polarization phases where, for example,  $|m_1| \neq |m_2|$  with  $|m_1|, |m_2| \neq 0$  or  $|n_1| \neq |n_2|$  with  $|n_1|, |n_2| \neq 0$  are allowed, but our energetic calculations have not found any of these partial polarization phases to be global energy minima for any values of  $n$ - $d$  parameters. We therefore believe that within our model partial spin and/or layer polarization phases are not stable ground states for any values of the parameters. We obtain our restricted HFA phase diagram by minimizing  $E^{\text{HF}}(m_1, m_2; n_1, n_2)$  with respect to the order parameters ( $m_1, m_2; n_1, n_2$ ) for each value of the system parameters ( $d, n$ ). Each ( $d, n$ ) point provides a unique set of ( $m_1, m_2; n_1, n_2$ ) which minimizes the HFA energy, and thus a complete phase diagram in ( $d, n$ ) parameter space is obtained, as shown in Fig. 2.

In Fig. 2 we show the results of our simple HFA phase diagram, which allows for only three phases  $S_0$ ,  $S_1$ , and  $A_1$ , with the  $A_0$  phase *not* stable at any values of the system parameters. Our calculated phase boundary (triangles in Fig. 2) between the high-density (i.e., low  $r_s$ ) paramagnetic bilayer phase ( $S_0$ ) and the low-density ferromagnetic bilayer phase ( $S_1$ ) occurs at a fixed  $r_s = 2.011$  (by contrast, the corresponding three-dimensional HFA ferromagnetic instability occurs at  $r_s = 5.45$ ) for all values of  $d$  because this is just the two-dimensional HFA instability to the formation of a ferromagnetic phase in which the Hartree energy does not play any role (note that interlayer correlations are being neglected in our approximation). At still lower (higher) density ( $r_s$ ) there is a transition (the phase boundary marked by squares) from the bilayer ferromagnetic phase ( $S_1$ ) to the monolayer ferromagnetic phase ( $A_1$ ) in Fig. 2 — this transition moves to lower densities (higher  $r_s$ ) as the interlayer separation  $d$  increases because of the higher cost in Hartree energy. Also

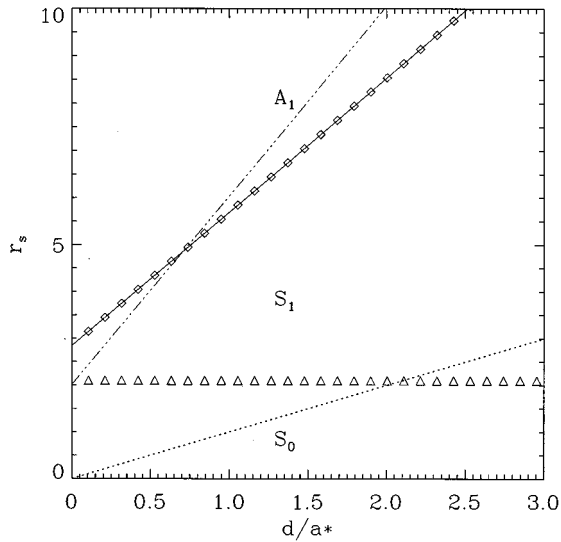


FIG. 2. Phase diagram for the restricted Hartree-Fock theory. The dotted line is the  $r_s = d/a^*$  line. The line with the three dots and a dash is the phase boundary of Ref. 11. The solid line is the phase boundary for spinless fermions.  $r_s = 1/a^*(\pi n)^{1/2} = 5.73759 \times 10^5 / (n)^{1/2}$  for  $n$  in units of  $1/\text{cm}^2$ .

shown in Fig. 2 are three lines. The lowest line (the dotted line) is the  $r_s = d/a^*$  line, which distinguishes approximately the regime of the average intralayer interelectron separation (i.e.,  $r_s$ ) being larger or smaller (the regime above or below the dashed line) than the average interlayer interelectron separation (i.e.,  $d/a^*$ ) in dimensionless units. The line with three dots and a dash in Fig. 2 is the phase boundary between the paramagnetic bilayer phase (below this line) and the paramagnetic monolayer phase (above this line), which is obtained if the spin polarization is ignored. We also show in Fig. 2 by a solid line the calculated phase boundary between the bilayer (below the line) and the monolayer (above the line) phases for *spinless* electron systems, where, by definition, the ferromagnetic spin polarized phases do not exist. Not surprisingly, the phase boundary (solid line) for spinless fermions coincides with the phase boundary (squares) separating the bilayer ( $S_1$ ) and the monolayer ( $A_1$ ) spin polarized ferromagnetic phases because the spin degree of freedom is frozen in the ( $S_1$ ,  $A_1$ ) spin polarized phases.

From Fig. 2 we conclude that within the restricted HFA any charge-transfer instability between bilayer and monolayer phases (we emphasize that fractional layer and/or spin occupancy states are not found to be ground states for any values of  $n$  and  $d$ ) in double-quantum-well systems must necessarily be preceded by a ferromagnetic phase transition and the charge-transfer instability (the squares in Fig. 2) is the  $S_1 \rightarrow A_1$  transition. Inclusion of interlayer correlation and tunneling effects should favor the bilayer phase over the monolayer phase, but our results show that, within the restricted HFA, there is indeed a low-density bilayer-to-monolayer charge-transfer instability.

### III. UNRESTRICTED HARTREE-FOCK THEORY

In this section, we study the double-layer spin-1/2 interacting electron gas in the unrestricted HFA. We show that

the predicted bilayer-to-monolayer charge-transfer transition at low electron densities (see the previous section) is an artifact of the restricted HFA resulting from treating the layer index as a classical Ising-like variable. Treating the layer degree of freedom in a fully quantum-mechanical fashion, we show in this section that, within the unrestricted HFA, the charge-transferred monolayer states are energetically unfavorable under any condition. This generic conclusion regarding the nonexistence of a charge-transfer instability is rigorously true in the HFA.

For convenience, we adapt the pseudospin description<sup>7,8,19</sup> for the layer degree of freedom, where  $\sigma_z = \pm 1$  represent the electronic states localized in the left or right layers, respectively, and  $\sigma_x = \pm 1$  represent the symmetric and antisymmetric states formed by the linear combinations of the left or right or  $\sigma_z = \pm 1$  eigenstates, respectively. In this language, population of all the electrons in a single layer corresponds to the pseudospin polarization in the  $\hat{z}$  direction, and population of all the electrons in the symmetric state corresponds to the pseudospin polarization in the  $\hat{x}$  direction. Thus, these two states (monolayer occupancy and symmetric state occupancy) are just pseudospin rotations of each other. In the HFA, the ground state of low-density electron systems tends to have complete spin and pseudospin polarizations in order to optimize the exchange energy. Since the Hamiltonian of an electron gas is spin-rotationally invariant [SU(2) symmetry], the energy of the system does not depend on the orientation of the spin polarization. The Hamiltonian of the double-layer system is, however, pseudospin-dependent [U(1) symmetry], so the energy depends on the orientation of the pseudospin polarization. As we will see shortly, the suggested bilayer-to-monolayer charge-transfer instability is an artifact arising from the classical restriction of the pseudospin polarization to the  $\hat{z}$  direction. The pseudospin rotated  $\sigma_x$ -polarized state necessarily has a lower energy than the monolayer occupancy  $\sigma_z$ -polarized state because there is no Hartree energy cost associated with charge transfer in the  $\sigma_x$ -polarized state.

To study the dependence of the ground-state properties on the orientation of the pseudospin polarization, we define the following orthonormal base in the pseudospin space:

$$\begin{aligned} |\xi\rangle &= \alpha|L\rangle + \beta|R\rangle, \\ |\bar{\xi}\rangle &= \beta|L\rangle - \alpha|R\rangle, \end{aligned} \quad (6)$$

where  $|L\rangle$  and  $|R\rangle$  represent the  $\sigma_z = \pm 1$  electronic states localized in the left and right layers, respectively,  $\alpha$  and  $\beta$ , with  $|\alpha|^2 + |\beta|^2 = 1$ , are the pseudospin rotation parameters determining a direction in the pseudospin space. Because of the symmetry of the system, we need only to consider the case where both  $\alpha$  and  $\beta$  are real numbers with  $1 \geq \alpha \geq \beta \geq 0$ . Our unrestricted HFA consists of doing the energy minimization with  $\alpha$ ,  $\beta$  as free parameters (with the constraint  $|\alpha|^2 + |\beta|^2 = 1$ ) whereas the earlier restricted HFA (Sec. II) made the specific choice of  $\alpha/\beta = 1/0$  (or  $0/1$ ).

We will examine the dependence of the HFA energy of the electron gas on the orientation of the pseudospin polarization, and compare it to that of an unpolarized (i.e., bilayer) state. For definiteness, we assume that there is no interlayer tunneling, since the effect of the interlayer tunneling

is always to oppose the charge-transfer instability. The ground state of the spin and pseudospin unpolarized phase (the  $S_0$  phase of Sec. II) is given by  $|S_0\rangle = \prod_k C_{k\xi\uparrow}^\dagger C_{k\xi\downarrow}^\dagger C_{k\xi\uparrow}^\dagger C_{k\xi\downarrow}^\dagger |0\rangle$ , where  $C_{k\xi s}$  ( $C_{k\xi s}$ ) is the creation (annihilation) operator for an electron with momentum  $k$ , pseudospin  $\xi$ , and spin  $s$ , and  $|0\rangle$  is the vacuum state. The corresponding HFA energy of the electron gas is

$$E_{S_0}^{\text{HF}} = \left( \frac{1}{r_s^2} - \frac{8\sqrt{2}}{3\pi r_s} \right) \frac{ne^2}{a^*}, \quad (7)$$

where  $r_s$  is related to the electron density through  $n = 1/(\pi a^{*2} r_s^2)$ . The ground state of the spin polarized but pseudospin unpolarized phase (the  $S_1$  phase) is given by  $|S_1\rangle = \prod_k C_{k\xi\uparrow}^\dagger C_{k\xi\downarrow}^\dagger |0\rangle$ . The corresponding HFA energy of the electron gas is

$$E_{S_1}^{\text{HF}} = \left( \frac{2}{r_s^2} - \frac{16}{3\pi r_s} \right) \frac{ne^2}{a^*}. \quad (8)$$

The ground state of the spin and pseudospin polarized phase is given by  $|P_\xi\rangle = \prod_k C_{k\xi\uparrow}^\dagger |0\rangle$ . The corresponding HFA energy of the electron gas is

$$E_{P_\xi}^{\text{HF}}(\alpha, \beta) = \left[ \frac{4}{r_s^2} + \frac{2d(\alpha^2 - \beta^2)^2}{ar_s^2} + \alpha^2\beta^2[I(0, r_s) - I(d, r_s)] \right] \frac{ne^2}{a^*}, \quad (9)$$

where

$$I(d, r_s) = \frac{4a^*}{d\pi} \int_0^1 dx x \int_0^\pi d\theta [1 - e^{-2\sqrt{2}d/a^* r_s (\sqrt{1-x^2}\sin^2\theta - x\cos\theta)}]. \quad (10)$$

As shown in Eq. (9), the energy of the spin and pseudospin polarized state explicitly depends on the orientation of the pseudospin polarization because the Coulomb interaction is layer index dependent. It is straightforward to show that the minimum of  $E_{P_\xi}^{\text{HF}}(\alpha, \beta)$  occurs when  $\alpha = \beta = 1/\sqrt{2}$ , i.e., when all the electrons reside in the symmetric state, and the maximum of  $E_{P_\xi}^{\text{HF}}(\alpha, \beta)$  occurs when  $\alpha = 1$  and  $\beta = 0$ , i.e., when all the electrons reside in a single layer. Thus, in the pseudospin space the monolayer occupancy phase is, in fact, an energy maximum for the possible pseudospin polarized states of the system. While both the symmetric state and the monolayer state optimize the exchange interaction energy by having complete pseudospin polarization, the symmetric state has on the average equal electron densities in the two layers and hence pays no cost in the static charging energy (the Hartree energy). The optimization of the exchange energy due to the pseudospin polarization is somewhat larger in the monolayer state than in the symmetric state because the intralayer Coulomb interaction is larger than the interlayer Coulomb interaction, but this difference is small compared with the Hartree energy cost for any values of the layer separation and electron density. Hence, the symmetric ( $\sigma_x$ -polarized) state is always energetically favored over the

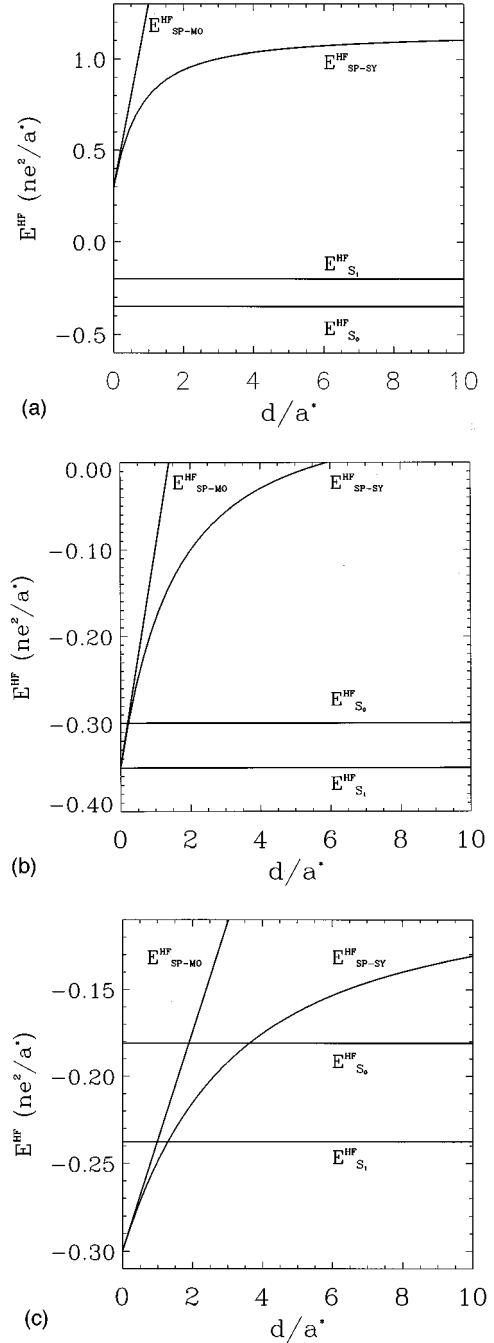


FIG. 3. The Hartree-Fock energies of a double-layer spin- $\frac{1}{2}$  interacting electron gas in the totally unpolarized state ( $E_{S_0}^{\text{HF}}$ ), in the spin polarized but pseudospin unpolarized state ( $E_{S_1}^{\text{HF}}$ ), in the spin polarized symmetric state ( $E_{\text{SP-SY}}^{\text{HF}}$ ), and in the spin polarized monolayer state ( $E_{\text{SP-MO}}^{\text{HF}}$ ) at different electron densities: (a)  $r_s = 2$ ; (b)  $r_s = 2(2)^{1/2}$ ; (c)  $r_s = 4(2)^{1/2}$ .

monolayer ( $\sigma_z$ -polarized) state. Note, however, that if the electrons were not charged objects so that there was no Coulomb charging energy involved in the charge-transfer instability, then exchange energy by itself is better optimized by the  $\sigma_z$  polarization and the monolayer state would be stable at low density.

In Fig. 3, we show the calculated HFA energies of the double-layer spin- $\frac{1}{2}$  interacting electron gas in the spin and pseudospin unpolarized state ( $E_{S_0}^{\text{HF}}$ ), in the spin polarized but

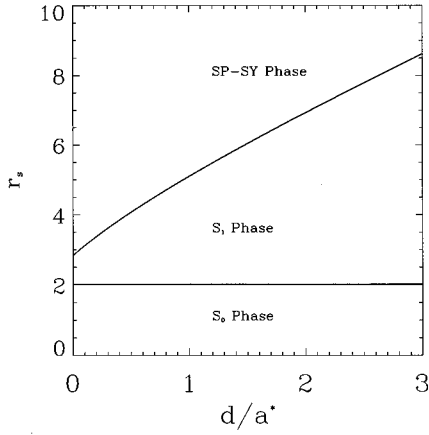


FIG. 4. Phase diagram of a double-layer spin- $\frac{1}{2}$  interacting electron gas in the Hartree-Fock approximation. There are three stable phases: the totally unpolarized phase ( $S_0$  phase), the spin polarized but pseudospin unpolarized phase ( $S_1$  phase), and the spin polarized symmetric phase (SP-SY phase). The charge transferred monolayer phase ( $A_0$  or  $A_1$  of Fig. 1) is not found to be a stable phase for any values of  $(r_s, d)$ .

pseudospin unpolarized state ( $E_{S_1}^{\text{HF}}$ ), in the spin polarized symmetric state [ $E_{\text{SP-SY}}^{\text{HF}} = E_P^{\text{HF}}(1/\sqrt{2}, 1/\sqrt{2})$ ], and in the spin polarized monolayer state [ $E_{\text{SP-MO}}^{\text{HF}} = E_P^{\text{HF}}(1, 0)$ ] as functions of the layer separation  $d$  at different electron densities. As mentioned above,  $E_{\text{SP-MO}}^{\text{HF}}$  is always larger than  $E_{\text{SP-SY}}^{\text{HF}}$ , hence, the bilayer-to-monolayer charge-transfer transition can never occur under any conditions. In the HFA, the energies of the pseudospin unpolarized states are independent of the layer separation because there is no interlayer direct or exchange interaction. On the other hand, the energies of the pseudospin polarized states are monotonically increasing functions of the layer separation. The ground state of the spin- $\frac{1}{2}$  double-layer system is found to be the spin and pseudospin unpolarized (paramagnetic bilayer) state at high electron densities, the spin polarized but pseudospin unpolarized (ferromagnetic bilayer) state at intermediate densities, and the spin polarized symmetric (ferromagnetic  $\sigma_x$ -pseudospin-polarized) state at low densities. The calculated unrestricted HFA phase diagram is shown in Fig. 4. There are three stable phases: the spin and pseudospin unpolarized phase ( $S_0$  phase), the spin polarized but pseudospin unpolarized phase ( $S_1$  phase), and the new spontaneously interlayer phase-coherent spin polarized symmetric phase (SP-SY phase). This phase diagram is similar to that of Fig. 2, except for one fundamental difference—the phase which exists at low densities and small layer separations in Fig. 4 is not the charge-transferred monolayer phase, but the pseudospin-rotated spin polarized symmetric phase where electrons on the average equally populate both layers. Inclusion of interlayer tunneling further reduces the energy of the SP-SY phase, making it even more energetically favored over the monolayer  $A_1$  phase. The spin polarization transition  $S_0 \rightarrow S_1$  is not affected by tunneling since the tunneling Hamiltonian is spin independent.

Our focus here is on the interesting spontaneous interlayer phase-coherent transition even in the absence of any tunneling energy. We emphasize that the transition from the  $S_1$

phase to the SP-SY phase in Fig. 4 is a true phase transition involving the spontaneous breaking of the pseudospin symmetry because it happens *even in the absence of any interlayer tunneling*. Without any interlayer tunneling (i.e., no spatial wave function overlap between the layers) the layer index is conserved in the Hamiltonian, and therefore the symmetric state, which is the even linear combination of the  $|L, R\rangle$  eigenstates, cannot be an eigenstate of the Hamiltonian unless there is a spontaneous breaking of the layer symmetry. An equivalent statement is that in the absence of tunneling one expects the ground state to be an eigenstate of the  $z$  component of pseudospin  $\sigma_z$ , *not* an eigenstate of  $\sigma_x$  as the symmetric state is. This is simply because the system Hamiltonian commutes (does not commute) with the  $\sigma_z$  ( $\sigma_x$ ) operator. In the presence of wave-function overlap (i.e., when interlayer tunneling is allowed) between the layers, the symmetric state is an allowed eigenstate of the system and is trivially the ground state of the noninteracting double-quantum-well structure. What is extremely interesting is our finding that even in the strict absence of interlayer tunneling, exchange interaction can drive the ground state of the system into the symmetric state (i.e., an eigenstate of  $\sigma_x$ ) at low electron densities. This is a surprising result because in the absence of tunneling all the terms in the Hamiltonian (the two-dimensional intralayer kinetic energy and the Coulomb interaction) conserve the layer index of an electron whereas the symmetry broken ground state turns out to be a coherent superposition of the electron being in the left and the right well state. In the strict absence of any tunneling, the low-density SP-SY phase of Fig. 4 is an example of spontaneous interlayer phase coherence.<sup>8</sup> We note that the layer density is not a good quantum number in the SP-SY phase even though there is no interlayer tunneling in the system. Thus the U(1) symmetry of the double-quantum-well Hamiltonian (without any tunneling) is broken spontaneously in the SP-SY phase. While being extremely interesting theoretically, the practical aspects of this spontaneous rotation in pseudospin space (from an eigenstate of  $\sigma_z$  to a symmetry-broken eigenstate of  $\sigma_x$ ) remain unclear because in the presence of any finite tunneling, the system should indeed be an eigenstate of  $\sigma_x$ . It is certainly possible to make double-quantum-well samples of high  $r_s$  and low  $d$ , which also have negligible interlayer tunneling (by having a very high potential barrier between the two layers). Our prediction is that such a system, if it is indeed in the SP-SY phase of Fig. 4, would behave as if it is in the (tunneling induced) symmetric state even though the actual tunneling matrix element is zero. The situation is analogous to a quantum Hall system<sup>8</sup> at the filling factor of 1, where it is believed that even in the absence of any Zeeman splitting there will be a spontaneous exchange-driven spin polarization transition. In our case, we have a spontaneous exchange-driven pseudospin polarization.

#### IV. LOCAL-DENSITY APPROXIMATION

There have been several recent experimental studies<sup>15–17</sup> searching for the charge-transfer instability. All these studies involve applying an external electric field (along the  $z$  direction) to continuously tune electron densities and then to measure layer electron densities via SdH oscillations. The experimental work involves<sup>15–17</sup> a GaAs- $\text{Al}_x\text{Ga}_{1-x}\text{As}$  double-

quantum-well structure (with an AlAs barrier layer) with an applied bias voltage between a front gate and the quantum wells. The action of this gate is to produce an electric field that is external to the device and to draw electrons from the quantum wells, thereby lowering the total electron density of the system. We study this system in the presence of an external electric field (and also in zero external field) using both the self-consistent local-density approximation (LDA) and the self-consistent local-spin-density approximation (LSDA) to determine the electron density in each well as well as the polarization state of the electron gas in each well, which can then be compared with the experimental results.

The basic idea behind the LDA for the spin unpolarized case is to self-consistently solve the coupled Poisson equation and the one-dimensional Schrödinger-like Kohn-Sham equation<sup>20</sup> in order to obtain the ground-state electron density of the quantum well. The LSDA is used to explore the possibility of a spontaneous spin polarization transition in the system. The LSDA is similar in spirit<sup>21,22</sup> to the aforementioned LDA. The major differences are that there are two Kohn-Sham equations (one for each spin component) that need to be solved in LSDA and that the exchange-correlation potential now depends<sup>22</sup> on both the electron density and the spin polarization of the electron gas. For the exchange-correlation potentials, we use the parametrization of Ceperley and Alder<sup>21</sup> for spin unpolarized or completely polarized and an interpolation formula due to von Barth and Hedin<sup>22</sup> for partial polarizations. Details of LDA (Ref. 18) and LSDA (Ref. 23) calculations for double-layer systems can be found in the literature.

Because the spin polarization of the final state can be affected by numerical inaccuracies, we perform the calculation using two very different initial values of polarization. One choice is a starting polarization that is small (10%) and the other is a starting polarization that is large (90%). If both choices lead to a polarized final state, then we say that state is polarized. If only one choice (say the 90% initial polarization) leads to a polarized final state, then we assume that the final result is affected by numerical inaccuracies and is uncertain. We follow the procedure of Eisenstein *et al.*<sup>18</sup> in allowing for the interlayer charge transfer in the presence of the external bias voltage within the LDA and LSDA. The external electric field is generated by adding additional charge to the topmost donor impurity sheet while maintaining overall charge neutrality between the donor sheets and the quantum wells.

We reproduce the results of Ying *et al.*<sup>16</sup> for two different double-quantum-well structures in the presence of an external electric field in Figs. 5(a), 5(b), and 5(c) finding good quantitative agreement between their results and ours. In Figs. 5(a) and 5(b) we calculate the front and back layer densities within the Hartree approximation (dashed line) and the LDA (solid line). The linear behavior of the electron densities within the Hartree approximation is expected as the electrons in the front layer attempt to screen the electrons in the back layer from the electric field. However, within the LDA, the density of the front layer decreases more quickly than in the Hartree approximation and the back layer density actually increases with increasing electric field. In Fig. 5(c), we show the results of the LSDA (three dots and a dash) showing that even with the possibility of spin polarized

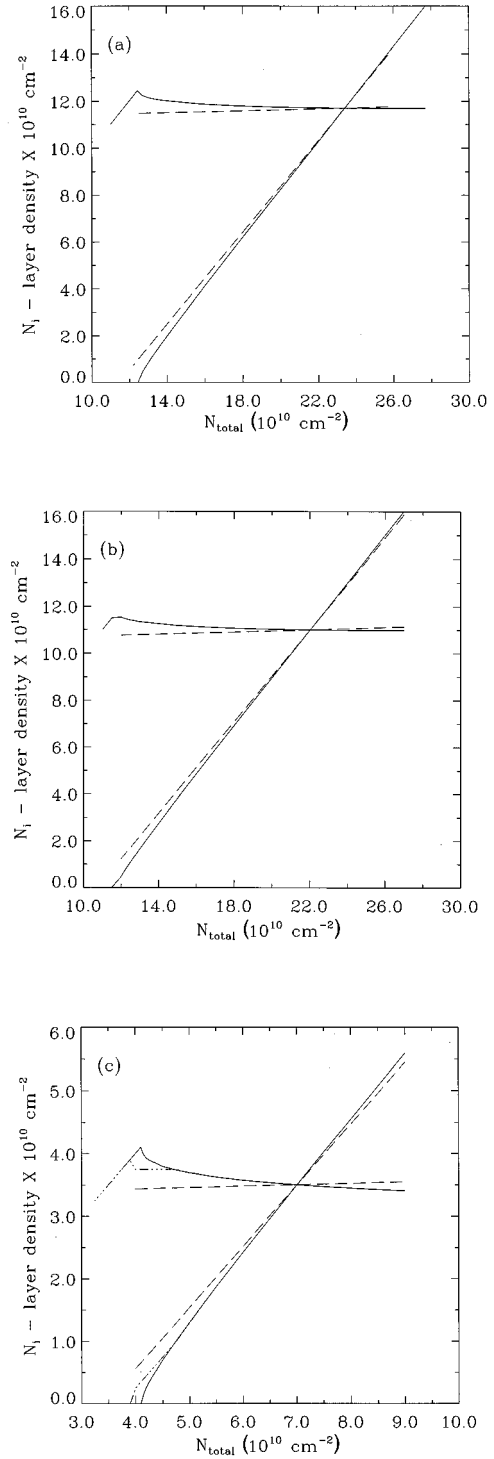


FIG. 5. (a) Plot of layer electron density versus total electron density for a double-quantum-well structure from Ref. 16 with a barrier width of 14 Å and well widths of 180 Å. The dashed line is the Hartree approximation. The solid line is the LDA. (b) Plot of layer electron density versus total electron density for a double-quantum-well structure from Ref. 16 with a barrier width of 70 Å and well widths of 150 Å. The dashed line is the Hartree approximation. The solid line is the LDA. (c) Plot of layer electron density versus total electron density for the double-quantum-well structure in (b). The dashed line is the Hartree approximation. The solid line is the LDA calculation. The line with three dots and a dash is the LSDA calculation.

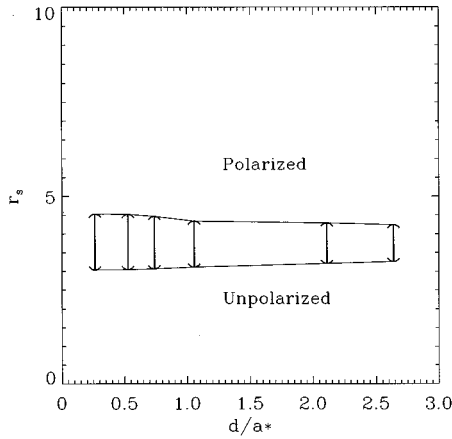


FIG. 6. Phase diagram of the double-quantum-well structure in Fig. 5(b) within the LSDA without an external electric field. The lower (upper) line corresponds to an initial spin polarization of 10% (90%) in the LSDA calculation. The region between these two lines is comprised of points for which the final spin polarization is dependent on the initial spin polarization used in the LSDA calculation.

states, the density of the back layer increases. This increase in the back layer density is a manifestation of the exchange-correlation induced “negative compressibility” effect,<sup>18</sup> which leads to a nonlinear layer depopulation in the external voltage.

We also perform the LSDA calculation on the same double-quantum-well structure as in Fig. 5(c) without an external electric field to calculate the ground-state phase diagram of the system. Our phase diagram, in electron density-layer separation space shown in Fig. 6, indicates that as the total density of the system is decreased, charge is not transferred from one well to the other, but instead, we find spin polarized ferromagnetic states (our  $S_1$  states of Fig. 2) for the electron gas in both wells. This establishes that although there is a net interlayer charge transfer *in the presence of an external electric field*, it is not the exchange-driven spontaneous bilayer-to-monolayer charge-transfer instability. We note that our LSDA phase diagram shown in Fig. 6 is qualitatively similar to the HFA phase diagram (e.g., Fig. 2) with two important differences: (i) The monolayer phases are not present, and (ii) the ferromagnetic transition occurs at a somewhat higher (lower)  $r_s$  (density) value, which is expected because the realistic LSDA calculation includes effects of finite well widths, etc., and includes correlation effects.

Our LDA and LSDA calculations establish that the external electric-field-induced interlayer charge-transfer experiments can be quantitatively understood as “negative compressibility”<sup>18</sup> effects. This point, in fact, has already been made in some of the experimental publications<sup>16,17</sup> where good agreement between the experimental data and the LDA calculations was shown to exist. Our predicted LSDA calculation based ferromagnetic spin polarization transition (Fig. 6) should occur at substantially lower densities than the experimental densities utilized in the existing literature.<sup>15–17</sup> Our predicted densities for the spin polarization transition in semiconductor double-well systems should, however, be accessible, particularly in hole-doped samples,<sup>24</sup>

where large effective  $r_s$  ( $\geq 25$ ) values have been recently achieved experimentally.

We emphasize that the experimental measurements carried out in the presence of an external electric field have little to do with the theoretical issue of a bilayer-to-monolayer charge-transfer instability because the application of the external electric field necessarily destroys the layer symmetry in the problem and the issue of a spontaneous symmetry-breaking (phase) transition or exchange instability becomes irrelevant. The situation is analogous (but *not* identical) to a magnetic transition in the presence of an external magnetic field, which is not a phase transition in any sense because there is an applied symmetry-breaking field. As mentioned before, the depopulation of subbands in the presence of an applied gate voltage is nonlinear due to exchange-correlation effects, and the so-called<sup>18</sup> exchange-correlation induced “negative compressibility” effect is the cause of the “unexpected” bump seen in the experimental results (see our Fig. 5 and Refs. 15–17), which is quantitatively explained by the LDA/LSDA calculations.

## V. CONCLUSION

We have obtained four theoretical results in this paper.

(i) We have shown that there cannot be any exchange-driven bilayer-to-monolayer charge-transfer instability in semiconductor double-quantum-well systems.

(ii) We have shown, within a mean-field HFA and also within a self-consistent LSDA theory, that there could be a ferromagnetic spin polarization transition in a double-quantum-well system at low (but accessible) densities.

(iii) We have shown that within a mean-field unrestricted HFA there is a quantum phase transition in a double-quantum-well system from a (spin polarized) bilayer state to a (spin polarized) interlayer phase-coherent symmetric state at low electron densities even in the absence of any interlayer electron tunneling—in the symmetric state the electron density in each layer develops spontaneous quantum fluctuations even though there is no overlap between the layer wave functions in the absence of tunneling.

(iv) We have shown that the experimental measurements<sup>15–17</sup> of layer or subband charge densities in double-quantum-well systems as a function of an applied external electric field can be understood quantitatively on the basis of LDA/LSDA calculations as arising from the two-dimensional “negative compressibility” effect.

Of these four results, obviously the most interesting are the results (ii) and (iii) above, both of which are based on reasonable but approximate theories. Our HFA phase diagrams invariably show low-density ferromagnetic phases where the electrons in each layer undergo a complete spin polarization transition. Our numerical LSDA calculation (Fig. 6) also finds the same result. While it is certainly possible (may even be likely) that our mean-field theory overestimates the density at which the ferromagnetic transition occurs, we believe that at high (low) enough  $r_s$  (density) the semiconductor double-quantum-well system does undergo a spin polarization transition. It would be difficult experimentally to directly observe this ferromagnetic transition because the actual spontaneous electronic magnetic moment associated with the spin polarized two-dimensional electrons is



rather small, and would be difficult to measure because of the large (orders of magnitude larger) background effect arising from the lattice. For the same reason, standard thermodynamic measurements (e.g., heat capacity) of the ferromagnetic phase transition may also be impossible. One possibility is to measure the two-dimensional Fermi momentum  $k_F$  in each layer, which will exhibit a jump by a factor of  $\sqrt{2}$  at the ferromagnetic transition. Transport measurements (in individual layers or in interlayer drag experiments) may be useful in this respect because, in principle, such measurements<sup>24,25</sup> are capable of indirectly measuring  $k_F$ . We believe transport<sup>24,25</sup> and capacitance<sup>18</sup> spectroscopies should show observable structures at the spin polarization transition in density sweep experiments.

Our most interesting theoretical finding is the possibility of a low-density quantum phase transition from a bilayer state to a coherent interlayer symmetric state (cf. Fig. 4), which happens even in the absence of any interlayer tunneling. Within the HFA we believe the existence of this phase transition to be rigorous. We speculate that this phase transition would exist even when correlation effects are included in the theory because correlation should affect the bilayer and the symmetric phase more or less equivalently. While being very interesting theoretically in its own right, a definitive experimental observation of this exchange-driven bilayer-to-symmetric phase transition (cf. Fig. 4) in semiconductor double-quantum-well systems would be difficult for a number of reasons. First, very low electron density ( $\sim 10^9$  cm<sup>-2</sup>) and low disorder double-quantum-well samples will be needed with rather large AlAs potential barriers to suppress tunneling. This is currently beyond the reach of MBE growth techniques for electron doped samples. It is, however, possible to make *p*-doped hole samples with very large effective  $r_s$  ( $\geq 25$ ) values,<sup>24</sup> which may be more suitable for observing our predicted spontaneous phase-coherent transition. Even if the desired samples are produced, few experiments (short of actual thermodynamic measurements which can look at specific heat anomalies at the phase transition) can actually distinguish between bilayer and symmetric states, because both states have the same *average* layer electron densities — in one case (bilayer) the layer electron density is an exact quantum number with no fluctuations while in the other case (symmetric) the layer electron density has quantum fluctuations and is not conserved. It seems that the interesting quantum phase transition shown in Fig. 4 may

remain only a tantalizing theoretical possibility in the near future.

A natural question arises about the nature of the quantum phase transition involved in the spontaneous breaking of the U(1) pseudospin symmetry in going from the bilayer  $S_1$  phase to the interlayer phase-coherent SP-SY phase in Fig. 4. Note that a similar phase transition has earlier been discussed in the literature<sup>8</sup> in the context of the quantum Hall effect phenomena in bilayer systems, where it has been argued that the U(1) symmetry of a double-quantum-well system in the absence of tunneling is spontaneously broken at a Landau level filling factor of 1 (and possibly at other filling factors as well). We find that there is nothing special about the quantum Hall situation in this context, and in fact as we show in this paper, a zero field exchange-induced spontaneous breaking of the U(1) symmetry is indeed possible at low densities. The theoretical phase diagram (Fig. 4) in the zero field situation is, in fact, richer because there are two tuning parameters ( $r_s$  and  $d$ ) controlling the phase transition (whereas in the quantum Hall case  $d$  is the only tuning parameter). Experimentally, of course, the situation is much more easily realized in the quantum Hall situation because it is much easier to obtain a Landau level filling factor of 1 than an  $r_s$  of 10. We speculate<sup>26</sup> that the nature of the U(1) pseudospin-symmetry-breaking phase transition in the zero magnetic field case is similar to that in the finite field quantum Hall situation,<sup>8</sup> even though further investigation of this issue in the zero field case is clearly warranted. While all the implications<sup>26</sup> of such a transition in our zero field case still remain to be worked out, it is likely that there is (at least the possibility of) an interesting finite temperature transition. In this context it may be worthwhile to point out that spontaneous interlayer phase coherence has been argued<sup>27</sup> to lead to interesting and observable effects in interlayer drag experiments.<sup>25</sup> We believe that such effects<sup>27</sup> would show up in the SP-SY phase as well and may be a way of identifying the new phase. More work is clearly needed in establishing the properties of the SP-SY phase and in elucidating the nature of the U(1) pseudospin-symmetry-breaking phase transition.

## ACKNOWLEDGMENTS

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