Novel Magnetic Instabilities in Semiconductor Double Quantum Wells

F. A. Reboredo and C. R. Proetto

Comisión Nacional de Energía Atómica, Centro Atómico Bariloche and Instituto Balseiro, 8400 Bariloche, Argentina

(Received 20 January 1997)

We present calculations of the electronic structure and total energy of double quantum wells for an arbitrary spin polarization of the system. This allows us to predict the order (first or second) of the magnetic phase transitions and the temperature range in which experiments should be done. We have found that, up to the present experimental situation, there is no contradiction with a local-density-approximation treatment. We have confirmed the existence of an antiferromagnetic phase and a ferromagnetic phase, but at lower densities than those claimed previously. Moreover, we have found a novel high-density ferrimagnetic phase under accessible electric fields. [S0031-9007(97)03589-8]

PACS numbers: 73.20.Dx

The local-density-approximation [1] of the density functional theory (DFT) [2] has been one of the most successful approaches to the many-body problem. The use of this method extends to the study of atoms, molecules, clusters, and solids [3]. In general the results obtained are not only qualitatively correct, but also quantitatively reliable with the additional advantage of a relatively easier implementation. During the last decades steady improvements on the experimental techniques have made possible the production of a great number of semiconductor artificial structures, which have also been studied theoretically in the frame of local density approximation (LDA). In particular $GaAs/Al_xGa_{1-x}As$ heterostructures have attracted intense attention because they are relatively easier to build, and the conduction band is particularly simple to describe. This latter feature implies that the effective mass approximation is very good [4], and also that one may use all the theoretical techniques developed for quantum many-body electron systems in an external potential, as LDA, time dependent LDA (TDLDA), spin-density LDA (LSDA), etc. In fact, these types of treatment have already been successfully applied to the description of ground and excited state properties of two-dimensional [5] and one-dimensional [6] electron gases, with very good agreement with experimental results. It should be pointed out that in the effective mass approximation the conduction electron band structure is replaced by a parabola (with an effective mass m^*) and simultaneously all the effects due to the core and valence electrons are taken into account via a dielectric constant (ε); besides, the epitaxial confinement is included in an effective external potential [see $V(z, \sigma)$ in Eq. (1) below]. With these approximations, which are known to be highly justified in these artificial heterostructures, the problem of the ground state of the remaining small density conduction electrons is in the same class of problems studied by DFT.

Recently, Das Sarma and Tamborenea [7,8] have predicted an antiferromagnetic phase (AP) transition to occur in double quantum well heterostructures. According to them, the transition arises as a result of the softening of the antiferromagnetic-spin-density excitation (ASDE) associated with the ground and first excited subbands of the double quantum wells (DQW). However, quite recent experimental results [9] show that the ASDE frequency does not go to zero when the density is reduced as predicted. This contradiction between theory and experiment was interpreted as a failure of LDA in the description of the ground state. Accordingly, it was claimed that a nonlocal approach was necessary to obtain good agreement with the experiments. This interpretation is somehow surprising because LDA is generally believed to lead to good results when the exact treatment of correlations is not a central issue; it is well known that the usual LDA fails in transition metals or in rare earths [3]. In this Letter we will show that, up to the present experimental situation, there is no contradiction with a LDA treatment, because the ASDE energy does not vanish when the density is reduced as it was claimed. However, the AP is still present but at smaller densities than those predicted in Refs. [7,8].

Our calculation is a generalization of the paramagnetic Kohn and Sham [1] equations to a spin polarized electron system [10]. Assuming translation symmetry of the system in the xy plane perpendicular to the growth direction z, the ground state is obtained solving a set of Schrödinger-like equations of the form

$$\left[-\frac{\hbar^2}{2m^*}\frac{\partial^2}{\partial z^2} + V(z,\sigma)\right]\phi_\sigma^m(z) = \epsilon_\sigma^m \phi_\sigma^m(z), \quad (1)$$

where m^* is the electron mass in GaAs, and σ is $+$ or $-$ for spins up and down, respectively. $\phi_\sigma^m(z)$ is the wave function corresponding to an electron with a spin projection σ and quantum number m, ϵ_σ^m being the corresponding eigenvalue. In Eq. (1) $V(z, \sigma)$ is the sum of the DQW potential $v_{DQW}(z)$, the Hartree potential $v_H(z)$, and the exchange-correlation potential $v_{XC}(z, \sigma)$. $v_{DQW}(z)$ is given by $-V_0[\theta(d_w + d_b/2 - |z|) - \theta(d_b/2 - |z|)]$, V_0 being the conduction band offset at the $GaAs/Al_xGa_{1-x}As$ interface, while $\theta(z)$ is the step function, d_w is the width of the GaAs wells, and d_b is the width of the $Al_xGa_{1-x}As$ barrier between the wells. The zero-temperature free electron density $\rho(z)$ is

given by

$$\rho(z) = \rho_+(z) + \rho_-(z)$$

$$= \frac{m^*}{2\pi\hbar^2} \sum_{\epsilon_\sigma^m < E_F} |\phi_\sigma^m(z)|^2 (E_F - \epsilon_\sigma^m), \qquad (2)$$

$\rho_+(z)$ $[\rho_-(z)]$ being the fraction of up (down) spin-polarized electrons and E_F the Fermi level of the system, which is determined by the charge neutrality condition: $\int_{-\infty}^{\infty} dz \rho(z) = N_S$, N_S being the two-dimensional delta-doping density. The Hartree potential (including the contribution of two positively charged δ-doped planes at coordinates $\pm h$ along z) is obtained solving the Poisson equation

$$v_H(z) = -\frac{2\pi e^2}{\varepsilon} \int_{-\infty}^{\infty} dz' |z - z'| \rho(z') + \frac{2\pi e^2 N_S}{\varepsilon} (\alpha z + h), \qquad (3)$$

where $-e$ is the electron charge, and ε is the dielectric constant of GaAs. The adimensional parameter α is adjusted to satisfy the boundary conditions: $\alpha \neq 0$ corresponding to either an asymmetric doping and/or to an external electric field. It remains to define the exchange correlation potential $v_{XC}(z, \sigma)$ which in the frame of LSDA is a function of the local density and the local spin density: $v_{XC}(z, \sigma) = v_{XC}(\rho(z), \sigma\eta(z))$, $\eta(z)$ being defined by $\eta(z) = \rho_+(z) - \rho_-(z)$. Following Refs. [7,8], we use a parametrization of the results obtained by Ceperley and Alder for fully polarized and unpolarized systems [11]. For intermediate polarizations we use an interpolation proposed by von Barth and Hedin [12] in which the correlation energy is assumed to have the same dependence as the exchange contribution. This is, however, a strong approximation and must be taken with care. Though the exchange and correlation contributions are both negative, they have opposite effects: The exchange contribution is maximum at $\eta(z) = 0$ where, on the contrary, the correlation energy is minimum (as one may expect because in a fully polarized electron gas the Pauli exclusion principle avoids interactions at short distances). For that reason any error in the exchange correlation energy, either accidental [13] or introduced in the parametrization [12], may produce important changes in the results. Equations (1)–(3) can be solved self-consistently using a standard procedure that will be explained elsewhere [14].

It is clear that if one imposes the paramagnetic condition $\eta(z) = 0$, $v_{XC}(z, \sigma)$, ϵ_σ^m, and $\phi_\sigma^m(z)$ will become independent of σ in the absence of magnetic fields; we will refer to them as the paramagnetic phase (PP) solutions $v_{XC}(z)$, ϵ^m, and $\phi^m(z)$ of the system. However, as soon as we allow $\eta(z)$ to be different from zero $v_{XC}(z, \sigma)$ becomes dependent of σ and consequently so do ϵ_σ^m and $\phi_\sigma^m(z)$ as well. This in turn may give rise to a difference $\eta(z) = \rho_+(z) - \rho_-(z)$ in the self-consistent solution.

In Refs. [7,8] the vanishing of the ASDE associated with transitions from the ground to the first excited subbands of the DQW ($\omega_{\mathrm{SDE}}^{01}$) was used as a signature of a magnetic instability from the PP towards the AP. It is clear that this criterion is valid when the transition studied is of second order; that is, when the energy of the PP is the absolute minimum of the system for $(\omega_{\mathrm{SDE}}^{01})^2 > 0$. It seems very difficult to decide whether this is correct or not, just from the knowledge of the paramagnetic ground state alone; accordingly, a better way to attack the problem is to start the self-consistent procedure from an initial state with a strong spin polarization. Eventually, the system may converge to a minimum different from the paramagnetic configuration. One may know which is the ground state just by comparison of the total energies of each phase that are given by the equation [10]:

$$\frac{E_G}{S} = \sum_{\sigma m} \int_0^{k_F^{\sigma,m}} \left(\epsilon_\sigma^m + \frac{\hbar^2 k^2}{2m^*}\right) \frac{k\, dk}{2\pi} - \int \frac{\rho(z) v_H(z)}{2} dz + \int [\rho(z)\epsilon_{xc}(\rho(z), \eta(z)) - \rho_+(z) v_{XC}(z, +) - \rho_-(z) v_{XC}(z, -)]\, dz, \qquad (4)$$

where $k_F^{\sigma,m} = [2m^*(E_F - \epsilon_\sigma^m)/\hbar^2]^{1/2}$, $\epsilon_{xc}(\rho(z), \eta(z))$ is the exchange-correlation energy per particle, and S is the system area. At zero temperature, the phase transition occurs when the energy gain per particle $\Delta E_{PP} = (E_G - E_G^{PP})/(SN_S) = 0$; E_G^{PP} being the value obtained for E_G in the PP. In Fig. 1 we compare the results of this procedure with those obtained from the condition $(\omega_{\mathrm{SDE}}^{01})^2 = 0$

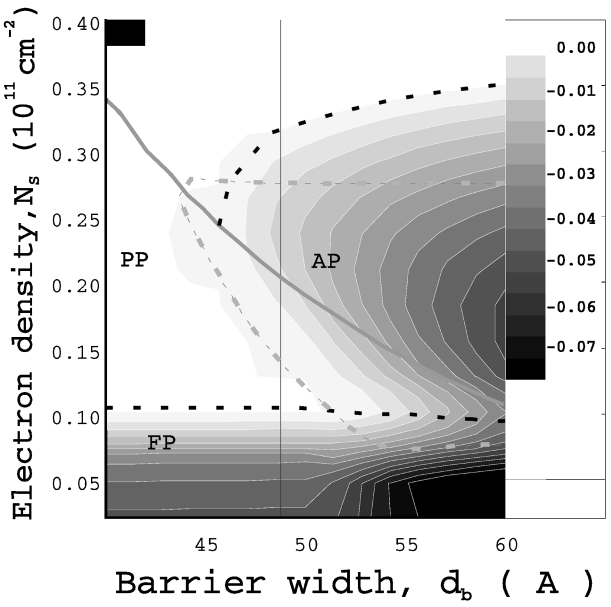

FIG. 1. Phase diagram of the DQW as a function of N_S and d_b. The shaded areas cover the regions where PP is expected to be unstable, the gray levels intensity (in meV/particle) being proportional to ΔE_{PP}. The dotted gray line shows the solution of $(\omega_{\mathrm{SDE}}^{01})^2 = 0$. The continuous line is the solution of $\epsilon^1 - E_F = 0$ in the PP, while dotted black lines mark the regions in which a first order transition is expected. The rectangle in the upper left corner indicates the size of the grid. The vertical line coincides with the vertical axis of Fig. 2.

used in Refs. [7,8], using the same set of parameters of these previous works ($V_0 = 220$ meV and $d_w = 139$ Å), setting $\alpha = 0$. The reader may note that the instability is moved to significantly lower density values from those predicted previously [13]. Let us define the following order parameters:

$$P_{\pm} = \frac{1}{N_s} \int_0^{\infty} dz[\eta(-z) \pm \eta(z)]. \qquad (5)$$

We will say that the system is in a FP (AP) when $P_+ \neq 0$, ($P_- \neq 0$), and $P_- = 0$ ($P_+ = 0$). We have identified three different phases in the system: At low densities the FP (ferromagnetic phase) is stable. As the density is raised the system passes through a first order transition to a PP for narrow barriers and to an AP for thick barriers. The AP is stable for intermediate densities and sufficiently thick barriers. There is a reentrance of PP in the low density regime. The low-density-transition AP $\rightarrow$ PP is of second order and the line predicted by both methods should be the same there. However, as $(\omega_{\mathrm{SDE}}^{01})^2$ is becoming quite small, the number of iterations needed to reach convergence diverges, and consequently, one needs increasingly long times to obtain similar results. This divergence is a clear signature of the order of the transition and there one may use the equation $(\omega_{\mathrm{SDE}}^{01})^2 = 0$. On the higher density side of the AP $\rightarrow$ PP transition, we obtain remarkably different results implying that it is a first order transition. We have verified numerically (introducing small fluctuations into the metastable phases) that the AP is metastable for even higher densities while the PP is metastable for lower densities up to the line defined by $(\omega_{\mathrm{SDE}}^{01})^2 = 0$. The order of the transition is also in correspondence with the behavior of P_+ and P_- (see below). From now on, we will use $\Delta E_{PP} < 0$ to define the unstable region. The FP includes a region at the large barrier width limit (black area) where all the electrons are essentially localized in the same well.

In Fig. 2 we summarize the dependence of the system on N_S and α for a fixed value barrier width; the rest of the parameters are the same as in the previous case. The symmetric case (vertical straight line in Fig. 1) corresponds to $\alpha = 0$, while $\alpha = 1$ corresponds to a DQW doped on a single side. When the reflection symmetry at $z = 0$ of the DQW is broken either by asymmetric doping or by an applied electric field [15], new interesting phenomena arise. In this case we have identified four phases: The low-density FP, the intermediate density AP, a high-density PP, and its reentrance below the AP, and a new high-density ferrimagnetic phase (FiP) which is absent at $\alpha = 0$. As all the particles of the system are affected in the phase transition one may roughly estimate from ΔE_{PP} that, even in the best conditions, the transition temperatures must be lower than 1 K.

In order to clarify the description given earlier, we will study the behavior of the system along the horizontal and the vertical straight lines of Fig. 2, which is shown in Figs. 3 and 4, respectively. In Fig. 3 we show the evolution of the system as a function of α for a fixed value of the density $N_S = 0.24 \times 10^{11}$ cm^{-2}. At $\alpha = 0$ in the AP, even though the wave functions $\phi_\sigma^m(z)$ depend on σ, the ϵ_σ^m are independent of σ, with only the states with $m = 0$ occupied. Besides, we have found that $\phi_+^m(z) = \phi_-^m(-z)$, and as a consequence [see Eq. (2)], $\eta(z) = -\eta(-z)$ and $\rho(z) = \rho(-z)$. Accordingly, $V(z, +) = V(-z, -)$, $P_+ = 0$, and $0 < |P_-| < 1$. Thus though the $v_{DQW}(z)$ and $v_H(z)$ have reflection symmetry around $z = 0$, for each component of the spin the exchange-correlation contribution is much stronger in one well than in the other; as a result, the ground sate wave functions $\phi_\sigma^0(z)$ are highly localized in opposite quantum wells. The increment in kinetic energy is compensated by the

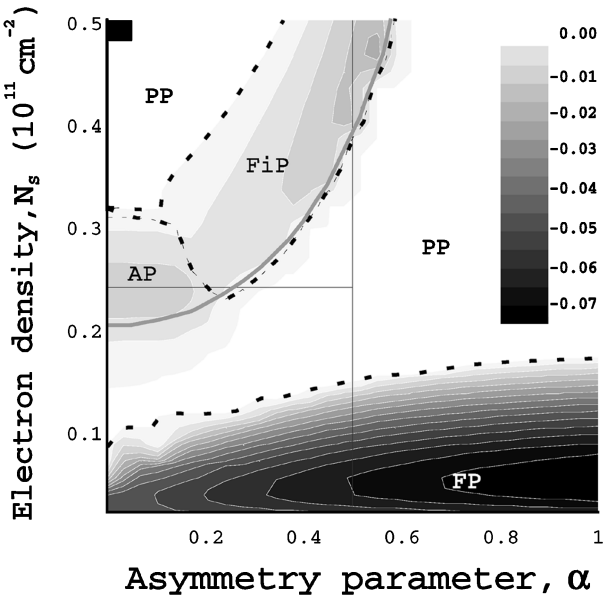

FIG. 2. Phase diagram of the DQW as a function of N_S and α, for $d_b \simeq 49$ Å. Same convention as in Fig. 1.

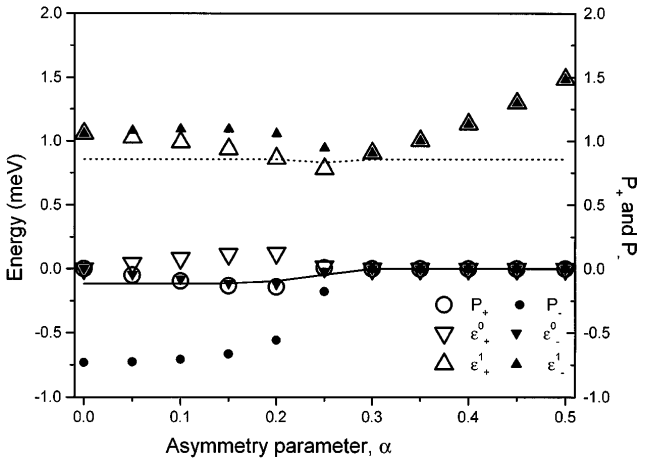

FIG. 3. Evolution of the subband structure as a function of α. The dotted line shows the values of E_F while the full line represent the values obtained for $\Delta E_{PP} \times 10$. The energy origin in the subband structure is placed at $(\epsilon_+^0 + \epsilon_-^0)/2$.

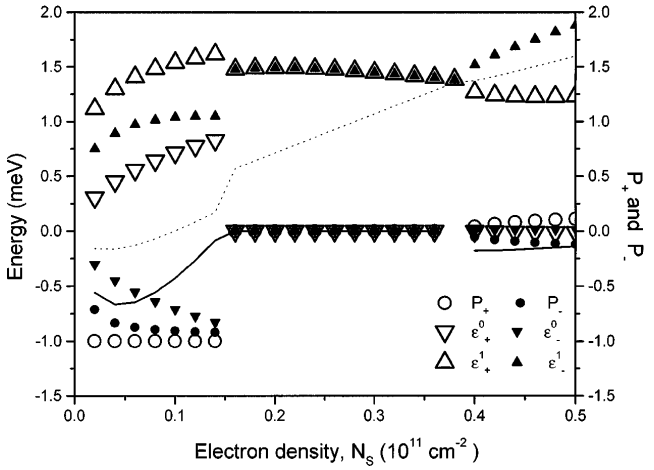

FIG. 4. Evolution of the subband structure as a function of N_S, for $\alpha = 0.5$. Same convention as in Fig. 3.

exchange-correlation energy gain. As soon as $\alpha \neq 0$, $v_H(z)$ becomes asymmetric and the degeneracy in ϵ_σ^m is broken. As α becomes larger, one component of the electron gas is increasingly allowed to tunnel towards the opposite well, which reduces $|P_-|$ significantly. Finally, both components become localized in the same well, $\eta(z) \to 0$, $P_\pm \to 0$, and the system converges to the PP. The behavior of the low-density FP (not shown) is also due to a combined effect of the Hartree and exchange-correlation contributions [14].

In Fig. 4 we present the results obtained for a fixed value of $\alpha = 0.5$, as a function of N_S. The low-density regime corresponds to the FP discussed above. In that region the electron gas is completely polarized [$\eta(z) = \rho(z)$ and $P_+ = -1$]; however, as $\rho(z)$ is highly asymmetric, P_- is different from zero. In fact, it is nearly equal to P_+, this means that $\eta(z) \simeq 0$ for $z > 0$. One spin component of the electron gas lowers its energy because of the exchange-correlation potential, while the other is pushed above the Fermi level. Accordingly, the density of states below the Fermi level is lower than in the PP, which in turn raises the kinetic energy of the FP. For low values of N_S, this kinetic energy cost is more than compensated by the exchange-correlation energy gain, but as the kinetic energy grows faster, there is a critical density N_S above which the paramagnetic phase is more stable and the system changes to a PP ($P_+ = P_- = 0$) in a first order transition. The existence of a new FiP for high densities can be understood as follows. For large values of α ($\alpha > 0.4$) the ground state wave function is highly localized in the lower energy QW, while the fist excited state is in the other. For intermediate values of N_S, E_F lies below ϵ^1, and the higher QW is empty, but as soon as $\epsilon^1 < E_F$ a small fraction of the total electronic density occupies the higher energy QW. In this part of the system, the electronic density is low enough to make a FP stable. As a result, the system is ferromagnetic in the higher QW and should be paramagnetic in the lower one, as the charge in it is much larger. However, the coupling between the wells induces a small polarization in the lower QW, $|P_+| \simeq |P_-|$ being a consequence of the fact that $\eta(z) \simeq 0$ for $z < 0$.

In summary, we have found that the antiferromagnetic transition predicted in Refs. [7,8] for double quantum wells occurs at smaller values of the density N_S and greater values of the interwell barrier d_b. This leaves the unstable region out of the range of almost all the samples used to confirm the phase transition [9]. We have also found that the transition is a first order one in its high-density boundary, and accordingly that the ASDE remains finite until the transition towards the AP occurs. We have studied the behavior of the system when its symmetry is broken and showed that a new ferrimagnetic phase appears in an asymmetric system at accessible higher densities (it is also in the range of electric fields that, as an additional advantage, can be obtained and tuned externally). Besides, we have estimated that the measurements must be performed at temperatures lower than 1 K to observe the transitions.

The authors would like to thank to P. Tamborenea for the general information about the current status of research on the subject, A. Pinczuk for sending us a copy of Ref. [9] prior to publication, and V. Grunfeld for a careful reading of the manuscript. One of us (F. A. R.) is indebted to CONICET of Argentina, for financial support.

[1] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).
[2] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).
[3] R. O. Jones and O. Gunnarsson, Rev. Mod. Phys. **61**, 689 (1989).
[4] G. Bastard, in *Wave Mechanics Applied to Semiconductor Heterostructures* (Les éditions de Physique, Les Ulis, France, 1988).
[5] T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).
[6] F. A. Reboredo and C. R. Proetto, Phys. Rev. B **55**, 1680 (1997), and references therein.
[7] S. Das Sarma and P. I. Tamborenea, Phys. Rev. Lett. **73**, 1971 (1994).
[8] R. J. Radtke, P. I. Tamborenea, and S. Das Sarma, Phys. Rev. B **54**, 13 832 (1996).
[9] A. S. Plaut *et al.*, Phys. Rev. B **55**, 9282 (1997).
[10] W. Kohn and P. Vashishta, in *Theory of the Inhomogeneous Electron Gas*, edited by S. Lundqvist and N. H. March (Plenum Press, New York, 1983).
[11] D. M. Ceperley and B. J. Alder, Phys. Rev. Lett. **45**, 566 (1980); J. Perdew and A. Zunger, Phys. Rev. B **23**, 5048 (1981).
[12] U. von Barth and L. Hedin, J. Phys. C **5**, 1629 (1972).
[13] Equations (30) and (31) of Ref. [8] are not correct since the coefficient of the exchange contribution is given in Rydberg (Ry) units, while the coefficients of the correlation contribution are in atomic units (2 Ry). We have found that this factor of 2 reduces significantly the critical density of the instability of the PP towards the AP.
[14] F. A. Reboredo and C. R. Proetto (to be published).
[15] A similar behavior is found by changing the width of one well with respect to the other.