Bilayer to monolayer charge-transfer instability in semiconductor double-quantum-well structures

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Disagreeing with the authors of the preceding Comment [F. A. Reboredo and C. R. Proetto, Phys. Rev. B **58**, 7450 (1998)] we contend that there is *no* exchange-correlation driven charge-transfer instability in semiconductor double-layer systems under any circumstances. The erroneous conclusion of the preceding Comment arises from its use of a *restricted* mean-field approximation within the LDA self-consistent scheme, which allows an energetic comparison *only* between the Ising-like bilayer and monolayer states. In a generalized *unrestricted* approximation more exotic phases are possible because the layer index is a quantum variable. Within the Hartree-Fock scheme, we prove a theorem that the spontaneous interlayer phase coherent state, called the SP-SY phase in our original paper [Phys. Rev. B **55**, 4506 (1997)], will always have a lower energy than the monolayer phase. The authors of the preceding Comment completely ignore this quantum interlayer phase coherent state. Thus any low-density layer instability in the double-quantum-well system, if it exists, will involve a transition from a bilayer to the interlayer phase coherent SP-SY state, and *not* to the monolayer state. The conclusion of the Comment arises from the *local* nature of the LDA scheme. [S0163-1829(98)07132-X]

In disagreement with the conclusion of the preceding Comment[1] we contend, reiterating our original conclusion[2] (and also in agreement with an independent recent finding[3]), that there is no exchange-driven low density transition from a bilayer to a monolayer state in semiconductor double-quantum-well structures. Below we explain the conceptual flaw in the Comment[1] and discuss its relevance to our original work.[2]

We obtained the following results in our original work:[2] (1) We established, both within the Hartree-Fock theory and the self-consistent LDA theory, that the ferromagnetic instability involving a transition to full spin polarization must always occur at a density higher than any charge transfer instability transition in a double-quantum-well structure. (2) We proved a theorem *within the unrestricted Hartree-Fock theory* that there cannot be any bilayer to monolayer charge-transfer instability because at low densities the interlayer phase-coherent state (the SP-SY phase in Ref. 2) must always have a lower energy than the monolayer state. (3) We calculated the electron charge-density profiles in the presence of an external electric field applied normally to the layers within the spin-polarized LDA self-consistent scheme, showing that the external field may cause rather sharp and nonlinear changes in the population densities of the individual layers (thus mimicking an apparent charge transfer ''instability,'' *but in the presence of an external electric field*). (4) We also carried out a zero-electric-field self-consistent spin-polarized LDA calculation to look for low-density spin and charge instabilities, finding a low-density transition to a ferromagnetic spin-polarized phase but *not* to a monolayer phase. It is important to emphasize that the preceding Comment[1] relates only to a part of the item numbered (4) above in our original work.

The disagreement between Refs. 1 and 2 is that in contrast to our LDA results reported in Ref. 2, in Ref. 1 a low-density charge-transfer instability is found to occur within the LDA self-consistent scheme in GaAs double quantum-well-structures. In spite of this apparent qualitative disagreement between the two LDA self-consistent calculations, there is in actuality *no real contradiction* between the two results: our LDA calculations in Ref. 2 looked for the convergent stable solutions of the Kohn-Sham equations whereas the results in the Comment[1] are obtained by comparing ground-state energies of the monolayer and the bilayer states. It is certainly possible for both sets of LDA results to be ''correct''—one[2] providing a metastable energy minimum and the other[1] presumably the global energy minimum (in the restricted subspace of monolayer and bilayer solutions only). The ''disagreement'' between the Comment[1] and our original work[2] is thus entirely superficial. We argue here that the numerical correctness of the restricted LDA self-consistent solution is totally irrelevant in answering the question posed in the title of our paper: The basic contention of our original work[2] is that a restricted mean-field calculation could lead to an erroneous conclusion about the existence of a charge-transfer instability because at low densities a completely different state, namely, the SP-SY state defined in Ref. 2, has a lower energy than the simple monolayer state considered in Ref. 1. It is therefore incumbent upon the authors of the preceding Comment[1] to show that the monolayer state that they find to be the ''stable'' ground state at low densities has a lower energy than the SP-SY phase of Ref. 2 before they can claim that they have established the existence of a charge-transfer instability in semiconductor double quantum-well systems.

To quantify our discussion let us write the ground-state energy, E, of the double-quantum-well system as a sum of several different contributions:

$$E = E_k + E_t + E_h + E_x + E_c , \tag{1}$$

where E_k (E_t) is the two-dimensional intralayer (interlayer hopping) kinetic energy contribution, E_h is the Hartree energy contribution arising from any possible charge imbalance in the system (e.g., the monolayer phase), and E_x (E_c) is the

exchange (correlation) contribution (both intralayer and interlayer) to the ground-state energy. It is well known that of these five contributions to the system energy only the exchange energy (E_x) favors a bilayer to monolayer charge transfer instability with all the other terms (E_k, E_t, E_h, and E_c) opposing the transition.

We showed in our original work[2] that neglecting correlation ($E_c=0$) and interlayer tunneling ($E_t=0$) effects the ground-state energy always (for all densities and layer separations) satisfies one of the two inequalities:

$$\text{high density: } E(\text{bilayer}) < E(\text{SP-SY}) < E(\text{monolayer})$$

$$\text{low density: } E(\text{SP-SY}) < E(\text{monolayer}) < E(\text{bilayer}), \tag{2}$$

where $E(x)$ indicates the energy for the state defined by x. (Note that if the state SP-SY is ignored in carrying out the energy comparison, one would erroneously conclude that the monolayer state is the true ground state at low densities with the *incorrect* inference of a charge-transfer instability.) Note that the Hartree-Fock theory ($E_t=E_c=0$) considered in Ref. 2 is the situation *most favorable* to the existence of a charge transfer instability because it leaves out two terms (E_t,E_c) opposing the monolayer state. *The fact that our theorem for the nonexistence of a bilayer to monolayer charge-transfer instability in semiconductor quantum-well systems was proven for the situation* ($E_t=E_c=0$) *most favorable to the monolayer state is definitive in ruling out the possibility of the monolayer state ever being the true ground state (in zero external electric fields) of the system.*

To make our discussions concrete we show in Fig. 1 our calculated phase diagram[4] within the *restricted* mean-field theory including approximate effects of E_t and E_c in the theory (and by keeping only four possible states in our restricted subspace: S_0, the spin-unpolarized paramagnetic bilayer state; S_1, the spin-polarized ferromagnetic bilayer state; A_0, the spin-unpolarized paramagnetic monolayer state; and A_1, the spin polarized ferromagnetic monolayer state), which goes beyond the exchange only—no tunneling ($E_c=E_t=0$) Hartree-Fock approximation. We include effects of E_c and E_t in our theory by using respectively the best available Monte Carlo estimate[5] for the correlation energy and by using a first-order perturbation theory in the interlayer hopping matrix element within the delta function potential well approximation[6] for the double-quantum-well structure. The important point is that we obtain a phase diagram qualitatively very similar to that given in the preceding Comment.[1] We are, therefore, perfectly willing to concede that an energy comparison between the monolayer and the bilayer state leads necessarily to the conceptually erroneous conclusion that there is a low-density charge-transfer instability. Thus, *any restricted* mean-field theory would mistakenly conclude that there may be a low-density charge-transfer instability.

It is reasonably obvious why the charge-transfer instability predicted by the restricted mean-field theory never happens in reality. Because the layer index is a quantum index (''pseudospin''),[7] the actual possibilities for it are much richer than the trivial classical (Ising-like) monolayer/bilayer possibilities considered in the preceding Comment.[1] In particular, at low densities a rotated pseudospin eigenstate,

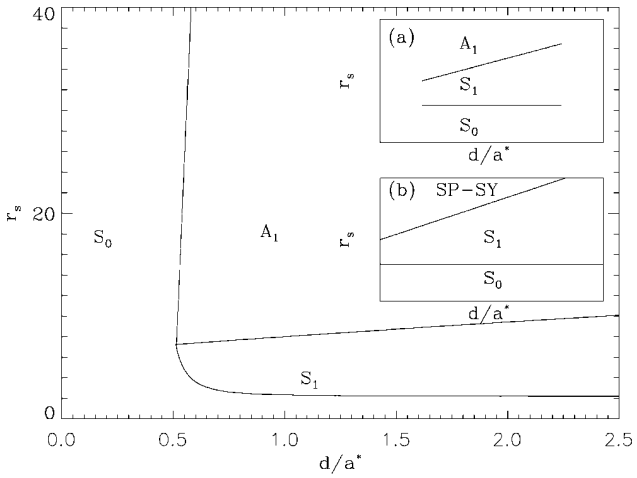

FIG. 1. Phase diagram for the *restricted* Hartree-Fock theory including (approximately) the effects of tunneling and correlation energy. This approximation shows a bilayer to monolayer charge transfer instability (for low electron density and large layer separation) due to the fact that the layer index has been treated as a classical Ising-like variable. Inset (a): Phase diagram for the *restricted* Hartree-Fock theory with $E_t=E_c=0$. The layer index has again been treated as a classical Ising-like variable. A bilayer to monolayer charge transfer instability is again found. Inset (b): Phase diagram for the *unrestricted* Hartree-Fock theory (i.e., allowing for arbitrary pseudospin polarization). The charge transferred monolayer phase is *not* found to be a stable phase for any values of (r_s,d). Here the layer index has been treated quantum mechanically. In all the figures r_s is the average planar electron separation measured in units of the effective Bohr radius a^* and d is the layer separation.

which is a linear symmetric (SY) combination of the two layer eigenstates, becomes energetically favorable over the simple monolayer state because this SY state has no Hartree energy cost and optimizes the exchange energy almost as well as the monolayer state does. The SY phase (in the absence of tunneling) is an example of a nonlocal interlayer spontaneous phase coherent state,[8] which exists even for $E_t \equiv 0$. Thus the system can lower its energy beyond the monolayer state (at low densities) by spontaneously going to this SP-SY state directly from the bilayer state. Note that the *average* electron density in the bilayer and the SP-SY states is indistinguishable. For more details on the SP-SY phase, we refer to Ref. 2. However, a manifestly *local* theory such as the LDA cannot, by definition, include a nonlocal phase-coherent state such as the SP-SY state, and therefore the conceptual error made in Ref. 1 cannot be fixed within the LDA scheme. A quantum Monte Carlo calculation,[3,5] which takes into account nonlocal *interlayer* phase coherent and correlation effects, may be able to obtain the complete phase diagram in the future. (We expect correlation corrections to the bilayer and the SP-SY phases to be very similar in magnitude.) Obviously, finite widths of quantum wells, etc., all act to oppose the charge-transfer instability, and once the theorem has been established for the Hartree-Fock case ($E_t=E_c=0$) other effects can only make the theorem stronger. We should point out[7] that there is a fundamental difference between the pseudospin and the spin of an electron, which is the key to understanding why a ferromagnetic spin polarization transition may occur in a double-quantum-well system

but *not* a bilayer to monolayer layer-polarization transition. This is because the electron-electron interaction Hamiltonian is independent of spin, but explicitly depends on pseudospin (i.e., the intralayer and interlayer interactions are different).

We conclude that the preceding Comment has a fatal conceptual mistake that cannot be fixed within the LDA scheme, which gives the incorrect result of there being a low-density charge-transfer instability in double-quantum-well systems. We also mention that experimental attempts to see a charge-transfer instability in semiconductor double-quantum-well systems have produced negative results[9] although the experimental samples are well into the monolayer regime of the calculated phase diagram in Ref. 1. It should perhaps be emphasized that LDA often fails to indicate phase transitions involving fundamental changes of symmetry in the problem—for example, Mott transition, Kohn-Luttinger superconductivity, charge-density wave, and Wigner crystallization. It is therefore not surprising that the authors of Ref. 1 fail to obtain the correct phase transition in the semiconductor double-quantum-well structures, and arrive at a conceptually wrong conclusion based on their LDA results. Any calculation that carries out a restricted mean-field calculation by comparing the energies of *only* the bilayer and the monolayer states may erroneously conclude that there is a low-density bilayer to monolayer charge transfer instability whereas in reality there is no such transition.

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[1] F. A. Reboredo and C. R. Proetto, preceding comment, Phys. Rev. B **58**, 7450 (1998).

[2] Lian Zheng, M. W. Ortalano, and S. Das Sarma, Phys. Rev. B **55**, 4506 (1997).

[3] S. Conti and G. Senatore, Europhys. Lett. **36**, 695 (1996).

[4] This result was taken off the published version of Ref. 2 to make the paper shorter (and because it was redundant in the sense that Ref. 1 is; it makes no statement qualitatively beyond the restricted Hartree-Fock result of Ref. 5).

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