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Steven G. Louie and Xuejun Zhu

Department of Physics
University of California

and

Materials Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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ELECTRON CORRELATIONS IN SEMICONDUCTORS: BULK COHESIVE PROPERTIES AND MAGNETIC-FIELD-INDUCED WIGNER CRYSTAL AT HETEROJUNCTIONS

Steven G. Louie and Xuejun Zhu

Department of Physics, University of California at Berkeley, and Materials Sciences Division, Lawrence Berkeley Laboratory, Berkeley, CA 94720, U.S.A.

Abstract

A correlated wavefunction variational quantum Monte Carlo approach to the studies of electron exchange and correlation effects in semiconductors is presented. Applications discussed include the cohesive and structural properties of bulk semiconductors, and the magnetic-field-induced Wigner electron crystal in two dimensions. Landau level mixing is shown to be important in determining the transition between the quantum Hall liquid and the Wigner crystal states in the regime of relevant experimental parameters.

1. Introduction

In this paper, we report recent progress on using a correlated wavefunction variational quantum Monte Carlo approach[1] in studying the properties of semiconductor systems. The approach employs a wavefunction of the Jastrow-Slater form and the exact Coulomb interaction between valence electrons. Two specific applications will be discussed. For bulk semiconductors, calculations have been carried out for the electronic and structural properties yielding results in excellent agreement with experiment. The method has further been applied to compute various quantities such as the electron pair correlation functions and single-particle orbital occupancy which are quantities not accessible in standard self-consistent-field theories.

For the two-dimensional (2D) electron system, we applied the approach to investigate the phenomenon of Wigner electron crystallization. Recent experimental evidence[2-4] indicate that this elusive crystal is perhaps finally observed in 2D electron and hole systems at semiconductor heterojunctions in the fractional quantum Hall regime. We calculated the ground-state energy of a triangular Wigner crystal as a function of carrier density, carrier mass, and applied magnetic field. The effects of exchange, correlations, and Landau level (LL) mixing are investigated. The transition from the incompressible quantum Hall liquid to the Wigner crystalline state is estimated by comparing the energy of the electron solid to that of the Laughlin state in various Landau level filling factors, υ. Our results show that LL mixing can significantly alter the critical filling factor υ_c for the liquid-solid transition. The relative importance of LL mixing is dependent on the density and effective mass of the carriers at a given Landau level filling factor.

In Section 2, a brief review of the present approach is given. Results on the cohesive properties of crystals are presented in Section 3, including results for the

electron pair correlation function. In Section 4, we discuss the magnetic-field-induced Wigner crystallization in 2D systems. Finally, a summary is given in Section 5.

2. Correlated Wavefunction Variational Quantum Monte Carlo Approach

The basic idea of the approach is to evaluate the ground-state properties of a system using the exact Hamiltonian but with a variational many-electron wavefunction and without further approximations. The Hamiltonian in our studies is of the form

$$H = \sum_{i=1}^{N} \left\{ \frac{\hbar^2}{2m} \nabla_i^2 + V_{ext}(r_i) + \frac{1}{2} \sum_{j \neq i} \frac{e^2}{r_{ij}} \right\} \tag{1}$$

where the last term is the exact Coulomb interaction between the electrons. For valence electron properties, the second term which describes the interaction of the electrons with the cores may be accurately approximated by norm-conserving ionic pseudopotentials.[1]

The many-electron wavefunction employed is of the form of an exponential correlation factor, the Jastrow factor, multiplying a Slater determinant of single particles:

$$\psi(r_1,\ldots,r_N) = \exp\left\{ \sum_{i=1}^{N} \chi(r_i) - \sum_{i<j} u(r_{ij}) \right\} D(r_1,\ldots,r_N) \tag{2}$$

where N is the number of electrons in the system. We retain both a single particle term χ and a two-particle term u. The two-particle term correlates the motion of the particles so that two electrons may avoid each other dynamically, and the one-particle term allows a relaxation of the charge density distribution in the presence of the two-particle term. In the calculations, variational parameters enter into χ and u. For accurate solid-state properties, simulation cell consisting of a piece of the crystal containing hundreds of electrons with periodic boundary conditions is required. Thus, it is necessary to use Metropolis Monte Carlo integration algorithms[5] in evaluating various physical quantities.

With a physically well-motivated variational wavefunction, the approach has proven to yield highly accurate ground-state energies and structural properties. In addition, a number of quantities which are not accessible to standard electronic structure methods have been obtained from the ground-state wavefunction. Included among these are the single-particle density matrix, the electron pair correlation function, and the occupancy for the single-particle orbitals.

3. Cohesive Properties of Diamond, Graphite and Silicon

With few exceptions,[1,6,7] present day *ab initio* electronic structure calculations on solids are based on self-consistent-field methods such as the local density functional formalism (LDA). These methods give excellent results for structural parameters and vibrational properties. However, in general, the SCF methods have problems in giving accurate absolute cohesive energies and in predicting properties of systems with highly correlated electrons (e.g., magnetic materials). The correlated wavefunc-

tion approach provides a means to treat electron interactions going beyond these standard *ab initio* methods.

Cohesive and Structural Properties. In our study of carbon- and silicon-based crystals,[1] simulation cells with periodic boundary conditions containing up to 216 electrons (or 54 atoms) were used. The Slater determinant part of the wavefunction is formed with single-particle orbitals obtained in a LDA calculation with *ab initio* norm-conserving pseudopotentials. A two-particle term of the form $u(r_{ij})=A[1-\exp(-r_{ij}/F)]/r_{ij}$ are used with A and F spin-dependent variational parameters. A single-particle term with one variational parameter which permits the valence charge density distribution to relax is also included.

The calculated energy of diamond is presented in Fig. 1 together with a fit to the Murnaghan equation of state. We obtained a calculated equilibrium lattice constant of 3.54 ± 0.03 Å and a bulk modulus of 420 ± 50 GPa in very good agreement with the experimental values of 3.567 Å and 443 GPa, respectively. Similarly accurate results for the structural parameters have been obtained for silicon. The calculated cohesive energies of diamond, graphite, and silicon are presented in Table I. The correlated wavefunction results are in excellent agreement with experiment[8]. In general, Hartree-Fock (HF) calculations significantly underestimate the cohesive energy whereas the LDA calculations tend to overestimate the cohesive energy, typically by 15 - 20% or more. Electron correlation effects, thus, play a very significant role in determining the crystal cohesive energy. For example, the valence electron correlation energy is calculated to be 4.1 eV per atom in diamond but only 2.4 eV for the isolated carbon atom.

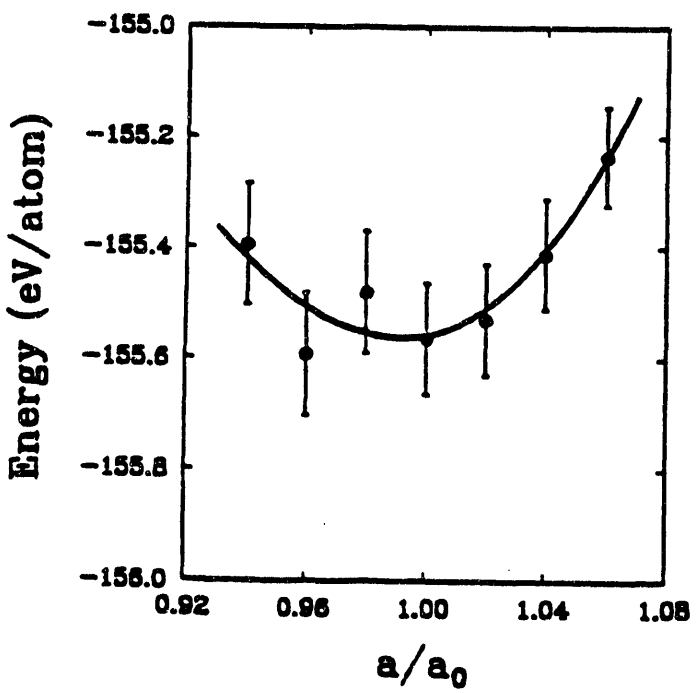

Fig. 1. Calculated energy of diamond (dots) together with a fit to the Murnaghan equation of state (line).

Table I. Calculated cohesive energies (in eV).

	LDA	VQMC	Experiment[a]
Diamond	8.63	7.45(7)	7.37
Graphite	8.65	7.40(7)	7.39
Silicon	5.29	4.81	4.7±0.1

a) Ref. 8

Electron Pair Correlation Functions. With the optimal wavefunction, it is conceptually straightforward to compute during the Monte Carlo walk various quantities such as the single-particle orbital occupation number, the quasiparticle excitation energies within a Feynman single mode approximation, and the pair correlation functions[9]. For example, the electron pair correlation function $g_{\alpha\beta}(r_1, r_2)$ may be evaluate through the relation

$$g(x_1,x_2)n(x_1)n(x_2) = N(N-1)\int |\psi(r_1 = x_1, r_2 = x_2, \ldots, r_N)|^2 dr_3 \ldots dr_N \qquad (3)$$

where n is the electron number density and spin indices are suppressed. Figure 2 illustrates the calculated pair correlation functions in diamond. Our results for $g_{\alpha\beta}$ of diamond and silicon show that, unlike the uniform electron gas case, g is indeed highly anisotropic and is a sensitive two-point function of r_1 and r_2. Both $g_{\uparrow\uparrow}$ and $g_{\uparrow\downarrow}$ are very rich in structure. For example, as seen in Fig. 2, the correlation hole, $h_{\uparrow\downarrow}(r) = g_{\uparrow\downarrow}(r_1,r) - 1/2$, for r_1 located at the bond center of diamond has a density distribution which is distinctly related to the structure and covalent character of the material. It is negative near r_1, the bond center, as expected, but $h_{\uparrow\downarrow}$ is positive only in the nearby low density antibonding/interstitial regions and not in the neighboring bond centers reflecting the covalent nature of diamond. Since the pair correlation function is intimately related to the exchange-correlation energy density, information such as those contained in Fig. 2 are valuable in a microscopic understanding of electron correlations in solids.

4. Magnetic-Field-Induced Wigner Crystals in Two Dimensions

Using the correlated wavefunction VQMC method, we have investigated[10] quantitatively the effects of exchange, correlations, and Landau level mixing to the ground-state energy of a triangular electron crystal in a perpendicular magnetic field.

The two-dimensional electron gas in a strong magnetic field, realizable experimentally in a MOSFET or a semiconductor heterojunction, exhibits a rich variety of phenomena.[2] By varying the carrier density, the effective mass, and the strength of the external magnetic field, a very intricate phase diagram is expected. The details of this phase diagram are only beginning to be explored experimentally and theoretically in recent years. Among the various possible phases, two prominent ones are the incompressible quantum Hall liquid phase and the Wigner crystal phase. The experimental evidence for the Wigner crystal phase are still being scrutinized. A reentrant behavior to an insulating phase near $\upsilon = 1/5$ for n-type samples and near $\upsilon = 1/3$ for p-type samples of GaAs/AlGaAs junctions of comparable carrier densities has been observed experimentally and interpreted as evidence for pinned Wigner crystals.

Electrons in Strong Magnetic Fields. Exchange-correlation effects in 2D in a strong magnetic field can be significantly different from those without the field because of the high degeneracy of the Landau levels.[11] For example, electron correlations resulting in lowering of the potential energy can occur without cost of kinetic energy in the strong field limit of fractionally occupied lowest LL. The presence of a strong B field introduces a new length, the magnetic length given by the radius of the Larmor orbit $\ell_B^2 = \hbar c/eB$, and a new energy, the cyclotron frequency $\omega_c = eB/m^*c$, into the problem. Both quantities are independent of the carrier density. Crystallization is

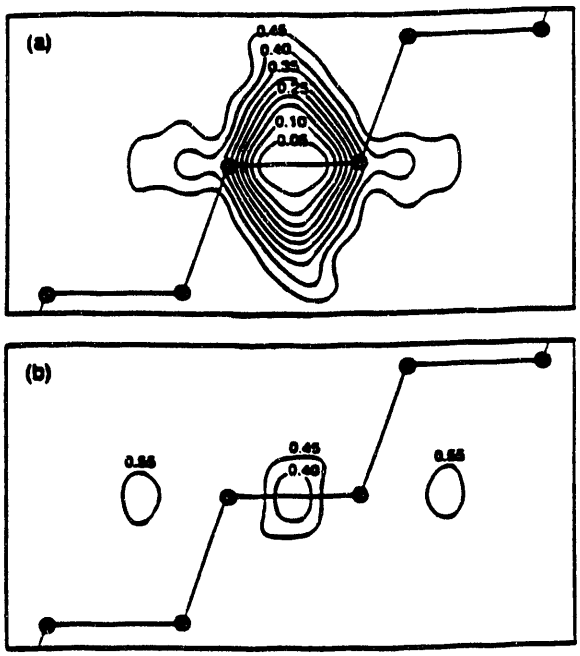

Fig. 2. Pair-correlation function in diamond for (a) parallel spin and (b) opposite spin with one electron at the bond center.

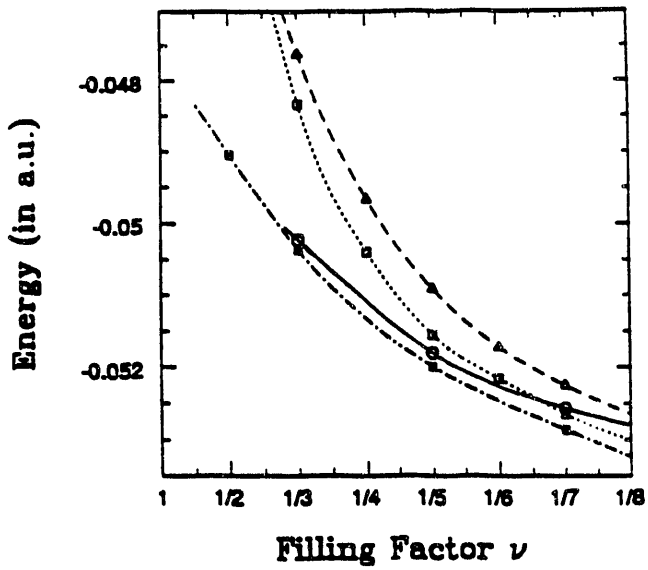

Fig. 3. Calculated Wigner electron crystal energy E - $\hbar\omega_c/2$ using exchange only with no LL mixing (---), correlations only with no LL mixing (···), correlations with LL mixing for $r_s = 20$ (-·-·). The energy of the Laughlin liquid (Ref. 15) is given by the solid curve.

expected when ℓ_B is less than the interelectron distance d, i.e., in the regime of $\upsilon < 1$. Landau level mixing is expected to be important when the electron-electron interaction energy $e^2/\varepsilon d$ is comparable or larger than the inter-LL spacing $\hbar\omega_c$. There are, therefore, three important materials parameters in determining the ground state, the density n, the effective mass m*, and the applied B field. The system is characterized by two independent dimensionless quantities: the filling factor υ which is a function of n and B and the electron gas parameter r_s which is a function of n and m*.

On the technical side, the usual periodic boundary conditions cannot be directly applied for electrons in a strong applied B field. This is because, although the field is uniform, the vector potential A is aperiodic, e.g., in the symmetric gauge A = (-yB/2, xB/2). The appropriate translational symmetry may be recovered by introducing the concept of rational fields[12] and noting that the physics of the system should be gauge invariant and that a finite translation in the present case is equivalent to a gauge transformation. These symmetry properties allow us to perform simulation with periodic boundary conditions in a magnetic field.

Our trial wavefunctions for the Wigner crystal are still in the general Jastrow-Slater form of an exponential correlation factor multiplying a function of the single-particle orbitals. The single-particle orbitals are chosen to be Gaussians localized about the lattice sites:

$$\phi_j(r) = \frac{1}{\sqrt{2\pi}} \frac{1}{\ell_B} \sum_{\{T\}} \exp\left\{-\frac{\beta}{4\ell_B^2}(r - R_j - T)^2\right\} \exp\left\{\frac{i}{2\ell_B^2}\left[x(R_j^y + T^y) - y(R_j^x + T^x)\right]\right\} \quad (4)$$

Here β is a variational parameter involving LL mixing. For $\beta = 1$, Eq. 4 corresponds to a wavefunction in the lowest LL localized on a lattice site R_j with the proper phase factors resulting from the use of a symmetric gauge at the origin. (The T's in Eq. 4 are translation vectors of the simulation cells associated with the periodic boundary conditions used.) All calculations reported here are done for spin-aligned electrons with a 100-electron simulation cell.

Results without Landau Level Mixing. We first discuss the results from wavefunctions which are made up of functions only from the lowest Landau level with either exchange or correlations included and then discuss in the next subsection the effects of LL mixing. Because of calculational difficulties, most previous work[13] was done with the assumption that LL mixing can be neglected. This portion of our work, thus, serves as a confirmation of previous work and to establish the validity of the present VQMC method. Our exchange-only (or HF) results are obtained by using a wavefunction which is just a Slater determinant of the single-particle orbitals given above with $\beta = 1$. The correlation-only results are calculated for distinguishable particles (again, with $\beta = 1$ for the orbitals) with the Jastrow correlation factor derived from the consideration of the magnetophonons similar to the work of Lam and Girvin.[14] In the large field limit, this Jastrow factor may be written as an analytic function of the complex variables which describe the position of the particles in 2D. Hence, in both calculations, there is by construction no mixing in of higher LL's into the ground-state wavefunction, and the kinetic energy per electron is trivially given by $\hbar\omega_c/2$.

The exchange-only and correlation-only results with no LL mixing are presented in Fig. 3 as the dashed and dotted curves, respectively. Previously, the most accurate energies for the 2D electron system in a strong magnetic field in the exchange-only approximation were calculated from the viewpoint of charge density waves (CDW)[13] rather than the explicit Wigner crystal wavefunction used here. Our present HF results are extremely close to the CDW results. Thus, little is gained by the explicit self-consistency procedure used in the CDW approach.

Our correlation-only results with no LL mixing are also in agreement with the results of Lam and Girvin[14] who used a special k-point sampling scheme in calculating the energy rather than the VQMC method. In this approximation, the Wigner crystal is considered as distinguishable particles correlated by the magnetophonons. Thus, although it contains correlation effects, exchange interaction is ignored. As noted in Ref. 14, at $\upsilon = 1/2$, the correlation-only result is, in fact, higher in energy than the HF result due to the rather large exchange interaction at this filling factor. However, a comparison of the Hartree, the exchange-only, and the correlation-only energies in the lowest LL approximation shows that correlation effects dominate over exchange in the range of $\upsilon \lesssim 1/3$. The energies for the Laughlin state taken from Ref. 15 are given by the solid curve in Fig. 3. The cross-over from the liquid state to the solid state is $\upsilon_c \approx 1/6.5$ as found by Lam and Girvin.

Effects of Landau Level Mixing. Our work here is motivated by the recent experimental work on the 2D hole systems which indicates the occurrence of a Wigner crystal phase around $\upsilon = 1/3$. This earlier transition from that of the electron case has been ascribed to a larger LL mixing due to the heavier hole mass in GaAs.

Even for the electron systems, the electron interaction energy is comparable to the cyclotron frequency. Thus, a quantitative understanding of LL mixing effects is important.

Because of the Coulomb repulsion between electrons on neighboring sites, it is energetically favorable to have a charge density distribution more localized than the one given by single-particle orbitals in the lowest Landau level. This is the driving mechanism for Landau level mixing. We put in this effect by optimizing the energy with respect to the parameter β in Eq. 4. Deviation of β from 1 corresponds to admixture of higher LL's into the wavefunction, and one needs to evaluate both the kinetic and interaction energies. The changes in energy and in the localization of the electrons due to LL mixing can be quite large in the range of $\upsilon = 1/2 - 1/5$ depending on the value of r_s. For example, at $\upsilon = 1/3$, with LL mixing, we find an increase in density at the lattice site by $\delta\rho(0)/\rho(0) = 70\%$ and an lowering in energy by $\delta E/E = -6\%$ at $r_s = 20$ whereas at $r_s = 2$ the same two quantities are 10% and -1%, respectively. Similar trends have been found in previous work[16] by including LL mixing effects perturbatively in the HF-CDW state or in small system calculations.

Our results with LL mixing and magnetophonon correlations for the case of $r_s = 20$ are depicted in Fig. 3 as the dashed-dotted curve. The parameters used in the calculation correspond roughly to those in the experiment on the 2D hole systems.[4] We see that there is a significant lowering in energy by allowing LL mixing in the range of υ of experimental interest. At $r_s = 20$, LL mixing effects are in fact larger than those of intralevel exchange-correlation effects. Compared to the energy of the Laughlin state, our results indicate within the approximations used here a crossover from the Laughlin state to the solid phase at near $\upsilon = 1/3$ and $\upsilon = 1/6$ for $r_s = 20$ and $r_s = 2$, respectively. A definitive comparison of the energy of the two phases cannot be made at this time because the Laughlin wavefunction[17] does not include higher LL's. However, one does not expect LL mixing to affect the liquid phase as much as the solid phase. Our results, thus, give quantitative support to the observation that, because of LL mixing effects, 2D hole systems at GaAs/AlGaAs interface crystallize at a larger υ than 2D electron systems at comparable densities.

5. Summary

A correlated wavefunction variational quantum Monte Carlo approach to electron-electron interactions in solids is reviewed. Highly accurate cohesive and structural properties, as well as quantities such as the electron pair correlation functions, have been obtained for carbon- and silicon-based crystals. The energetics of the magnetic-field-induced Wigner crystal in 2D in the fractional quantum Hall regime has also been investigated. Landau level mixing effects are shown to be significant in the range of density and magnetic field strength of experimental interest. Our results provide an explanation for the recent observation of a re-entrant insulating transition near $\upsilon=1/3$ for 2D hole systems at GaAs/AlGaAs interfaces.

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