Frozen electron solid in the presence of small concentrations of defects

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We investigate the freezing of a low-density electron liquid for a two-dimensional electron layer in the presence of low levels of defects typical of a high-quality semiconductor interface. We use a memory function approach with mode-coupling approximation and include the effect of strong electron-electron correlations, which we find are crucial for the transition. For a range of low impurity concentrations we find a stable frozen solid with a liquidlike short-range order. At higher impurity concentrations the electrons localize separately and there is no short-range order. Our electron-density vs peak-mobility phase diagram at zero temperature is in agreement with recent metal-insulator transition experiments in silicon heterostructures. [S0163-1829(96)08935-7]

Numerical simulation studies of a two-dimensional electron liquid predict that it would only condense into a Wigner crystal at exceedingly low densities $r_s \gtrsim 37 \pm 5$.[1] However, the energy differences between the liquid and crystalline states are already very small for densities $r_s \gtrsim 10$. This suggests that low levels of defects might be sufficient to induce a transition to a solid phase at a much higher density than $r_s = 37$. Pudalov *et al.*[2] working with electron inversion layers at silicon metal-oxide-semiconductor field-effect transistor (MOSFET) interfaces, observed a collective metal-insulator transition at densities as high as $r_s \simeq 8$. The nature of the coherent insulating state is still not fully clear.

We have found that low levels of disorder typical of those in state-of-the-art semiconductor substrates acting in concert with strong correlations between electrons can cause a transition to a coherent solid of localized electrons at densities as high as $r_s \simeq 7$. The levels of disorder needed for this typically correspond to far fewer impurities than electrons.

The solid is not a Wigner crystal but a frozen macroscopically coherent state with *liquidlike short-range order*. It is quite different from another frozen state obtained from localized electrons interacting with a disordered medium which has been termed an electron glass.[3–5] The vanishing of a soft Coulomb gap in the single-particle density of states or a nonzero value of the Edwards-Anderson–like order parameter provides the signature in that case.

Our mechanism for electron localization is also different from the mechanism discussed earlier for free-electron scattering from randomly distributed impurities.[6] There localization was obtained by increasing the strength of the impurity-potential fluctuations (see also the discussion in Ref. 7).

Our localization, in contrast, is mainly driven by the strong effect of electron correlations at large r_s. The mechanism for the transition is associated with the increasing relative size of the correlation hole surrounding each electron as r_s increases. From Ref. 1 we know that for $r_s \gtrsim 10$ the exchange correlation hole excludes all other electrons from a central region surrounding it as if the electron had a hard core. For $r_s \gtrsim 10$ the electrons with their exchange correlation holes resemble hard disks. With decreasing electron density the fraction of the total area occupied by excluded regions approaches the close packing limit[8] and at this stage it becomes increasingly difficult for electrons to pass by each other. A small amount of impurity disorder introduces pinning centers and breaks the translational invariance of the system. The localization is quasiclassical because it is driven by the increasing size of the exchange-correlation hole. We find that with a small amount of disorder included the electrons can freeze for $r_s \gtrsim 7$.

We investigate the transition to a nonergodic phase using a model of the glass transition originally constructed to account for the freezing of dense classical systems.[9,10] We have adapted this to a quantum system.

We study the limiting behavior of the Kubo relaxation function $\lim_{t\to\infty}\{\Phi(\boldsymbol{q},t) \equiv (N(\boldsymbol{q},t)|N(\boldsymbol{q},0))\}$, defined on the normalized density fluctuation basis, $N(\boldsymbol{q},t) = \rho(\boldsymbol{q},t)/\sqrt{\chi(\boldsymbol{q})}$, where $\chi(\boldsymbol{q})$ is the electron liquid static susceptibility. At lower electron densities the density fluctuation operators $\rho(\boldsymbol{q},t)$ are an appropriate choice for the dynamical variables of the system rather than single-electron wave functions since the shape of the exchange correlation hole is determined mainly by the strong repulsive interactions. Exchange effects play only a secondary role here and interference effects between single-electron waves are not expected to produce solidification.

In the liquid phase $\Phi(\boldsymbol{q},t)$ tends to zero for times greater than the macroscopic relaxation time of the system. When a freezing point is approached the time decay in $\Phi(\boldsymbol{q},t)$ becomes very slow and eventually stops, implying a complete arrest of density fluctuations. We use a memory function approach with mode-coupling approximation to investigate the existence of the transition.[9] Being a quantum system we are not able to look at dynamic properties of the liquid as the transition is approached since, unlike the classical case, there exists no unique relation between the dynamic structure factor and response function. However, even in the quantum case the relation does exist if the frequency is first set equal to zero, which enables one to write quantum-correlation

functions in terms of the relaxation function in the memory function. Thus by directly searching for a singularity in $\Phi(\boldsymbol{q},z)$ at $z=0$ we can use the approach to search for the transition in a quantum system.

Within the Mori-Zwanzig formalism[11] the exact equations of motion for the density-relaxation function can be expressed in terms of the relaxation function of the fluctuating forces $M(\boldsymbol{q},z)$. In the $z\to 0$ limit the expression reduces to,

$$\lim_{z\to 0}\{-z\Phi(\boldsymbol{q},z)\}\equiv f(\boldsymbol{q})=\left[1+\frac{\Omega(\boldsymbol{q})}{M(\boldsymbol{q})}\right]^{-1}. \tag{1}$$

$\Omega(\boldsymbol{q})=\boldsymbol{q}^2/[m^{\star}\chi(\boldsymbol{q})]$ and $M(\boldsymbol{q})\equiv-\lim_{z\to 0}zM(\boldsymbol{q},z)$. Nonzero values of the order parameter $f(\boldsymbol{q})$ signify that spontaneous fluctuations remain for an infinite time.

We approximate $M(\boldsymbol{q},z)$ using mode-coupling theory.[12] The contribution to the memory function from electrons interacting with the disorder is

$$M_{de}(\boldsymbol{q})=\frac{1}{m^{*}\boldsymbol{q}^2}\sum_{\boldsymbol{q}'}[n_i W_{ie}^2(\boldsymbol{q}')S_i(\boldsymbol{q}')+W_{\mathrm{sf}}^2(\boldsymbol{q}')](\dot{\boldsymbol{q}}\boldsymbol{q}')^2 \times\chi(|\boldsymbol{q}-\boldsymbol{q}'|)f(|\boldsymbol{q}-\boldsymbol{q}'|), \tag{2}$$

where n_i is the density of impurities and $S_i(\boldsymbol{q})$ the impurity structure factor. The electron-impurity potential is $W_{ie}(\boldsymbol{q})=[(2\pi Ze^2)/(\epsilon|\boldsymbol{q}|)]F_i(\boldsymbol{q})$ and the surface roughness scattering at the interface is $W_{\mathrm{sf}}(\boldsymbol{q})=\sqrt{\pi}\Delta\Lambda\Gamma(\boldsymbol{q})\exp[-(q\Lambda)^2/4]$. For the form factor $F_i(\boldsymbol{q})$ we use Eq. 4.28 in Ref. 13. Values for the surface roughness parameters $\Lambda=0.37$ nm and $\Delta=2.0$ nm are taken from experiments on silicon MOSFET's.[14] The expression for $\Gamma(\boldsymbol{q})$ is taken from Ref. 15. Götze[6] used an expression similar to Eq. (2) for the total memory function $M(\boldsymbol{q})$, taking for the electron-impurity interaction a model potential.

The contribution to the memory function from the mutual interaction between the electrons contains higher-order correlation functions, which we approximate by two independently propagating density fluctuation modes. We take the lowest-order coupling between these modes, which is the bare Coulomb interaction $V(q)$,

$$M_{ee}(\boldsymbol{q})=\frac{1}{2m^{*}\boldsymbol{q}^2}\sum_{\boldsymbol{q}'}[V(\boldsymbol{q}')(\dot{\boldsymbol{q}}\boldsymbol{q}')+V(|\boldsymbol{q}-\boldsymbol{q}'|)[\dot{\boldsymbol{q}}(\boldsymbol{q}-\boldsymbol{q}')]]^2 \times\chi(\boldsymbol{q}')\chi(|\boldsymbol{q}-\boldsymbol{q}'|)f(\boldsymbol{q}')f(|\boldsymbol{q}-\boldsymbol{q}'|). \tag{3}$$

The memory function $M(\boldsymbol{q})$ is the sum of the contributions $M_{eh}(\boldsymbol{q})$ and $M_{ee}(\boldsymbol{q})$. The quantum effects of the exchange-correlation hole enter our calculation through the static susceptibility $\chi(\boldsymbol{q})$. This is determined from numerical simulation data for the static structure factor[1] $S(\boldsymbol{q})$ which contains information on the structure of the exchange-correlation hole. We obtain $\chi(\boldsymbol{q})$ using the fluctuation-dissipation theorem and a static local-field approximation.[16]

The nonergodicity parameter $f(\boldsymbol{q})$ couples $M_{de}(\boldsymbol{q})$ and $M_{ee}(\boldsymbol{q})$. This leads to an interdependence between the electron-disorder scattering and electron-electron correlations. Equations (1)–(3) are solved self-consistently for $f(\boldsymbol{q})$. Nonzero solutions of these equations for $f(\boldsymbol{q})$ are shown in Fig. 1 for a range of impurity densities. We assume the impurities have no short-range order so $S_i(\boldsymbol{q})=1$.

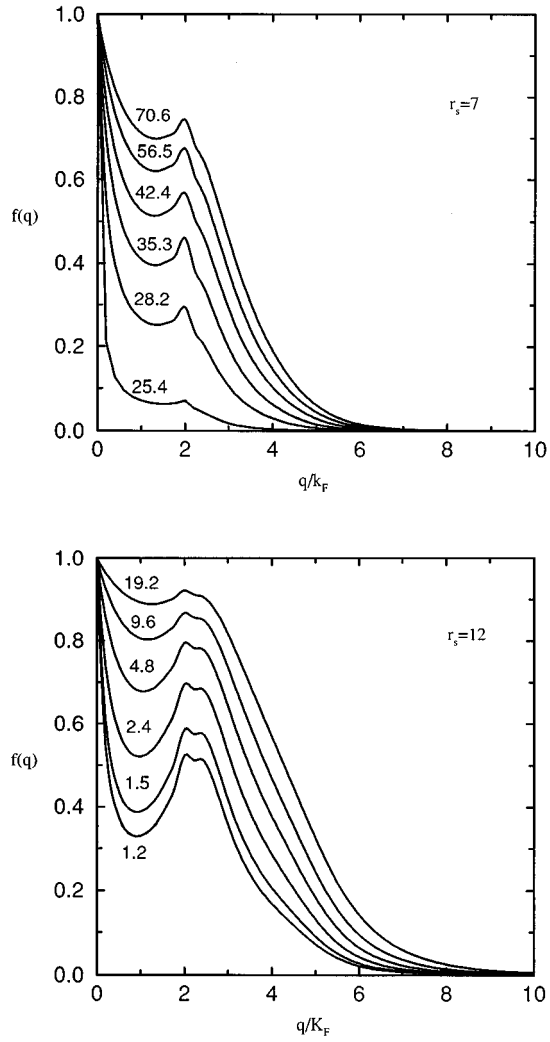

FIG. 1. Nonergodicity parameter $f(\boldsymbol{q})$ for electron densities $r_s=7$ and $r_s=12$. The labels are the impurity density n_i in units of 10^9 cm^{-2}.

At the higher electron density ($r_s=7$) shown in Fig. 1(a), the peak in $f(\boldsymbol{q})$ centered at $\boldsymbol{q}=0$ steadily broadens and its overall shape evolves into a Gaussian-like function that does not show evidence of short-range or long-range order. [The cusp in $f(\boldsymbol{q})$ at $|\boldsymbol{q}|/k_F=2$ only reflects the well-known cusp in the Lindhard function $\chi_0(\boldsymbol{q})$.] We find that $f(\boldsymbol{q})$ continuously diminishes as the disorder is decreased. However it remains nonzero down to the smallest disorder strengths. We conclude at this moderate electron density that the localization is noncoherent and that it is driven by the impurities. Noncoherent localization is the type to be expected at higher electron densities where electron-disorder scattering dominates over many-body electron-electron correlations. Recent electric transport experiments on Si MOSFET's (Ref. 17) have found non-coherent localization in the range of electron densities $5\lesssim r_s\lesssim 9$.

When, by lowering the electron density, we increase the importance of the many-body electron-electron correlations we find that the nature of the localization changes. This is illustrated in Fig. 1(b). At $n_i=1.20\times10^9$ cm^{-2} $f(\boldsymbol{q})$ undergoes a discontinuous transition. For $n_i>1.20\times10^9$ cm^{-2} $f(\boldsymbol{q})$ is nonzero for all $\boldsymbol{q}$. For $n_i<1.20\times10^9$ cm^{-2} $f(\boldsymbol{q})$ is

only nonzero in the immediate vicinity of $\boldsymbol{q}=0$. This is associated with weak localization which is expected in low-dimensional systems for low disorder. We use the discontinuous jump in $f(\boldsymbol{q})$ as the criterion to distinguish this transition from the noncoherent localization in Fig. 1(a).

In Fig. 1(b) the new structure in $f(\boldsymbol{q})$ centered around $|\boldsymbol{q}|/k_F \approx 2.4$ reflects a buildup of short-range order. The short-range order resembles the short-range order existing for the electron liquid at the same density. For this reason we identify this frozen phase with coherent localization. The electron-electron correlations are crucial for this phase. If we neglect them by using a Hubbard-like expression for $\chi(\boldsymbol{q})$ in Eqs. (2) and (3) we do not get the discontinuous jump in $f(\boldsymbol{q})$.

This phase persists until about $n_i = 1.8\times 10^{10}$ cm^{-2}. By then the peak in $f(\boldsymbol{q})$ around $|\boldsymbol{q}|/k_F \approx 2.4$ has practically disappeared which indicates loss of short-range order. This is to be expected when the impurity density is of the order of electron density since for such high impurity concentrations the localization should be incoherent.

The Lindemann ratio at the melting transition can be calculated from the probability distribution function $P(r)=2\pi r f(r)$ where $f(r)=(2\pi)^{-2}\int d\boldsymbol{q} e^{i\boldsymbol{q}\cdot\boldsymbol{r}} f(\boldsymbol{q})$. For $r_s = 12$ $P(r)$ at the melting point has a maximum for $r/r_0 = 0.29$. This is in good agreement with the melting of the Wigner lattice.[18]

Our coherent state is different from the electron solid discussed by Chui and Tanatar.[19] They found for a fixed random distribution of impurities that the lowest-energy state was a crystalline solid at density $r_s \simeq 7.5$. Their state became increasingly amorphous if the electron density was lowered further. The solid state discussed in Ref. 19 is a ground state and is thus different from our metastable frozen state. Our frozen electrons are in a nonequilibrium state. A hysteresis experiment could distinguish between these states.

In recent experimental work Pudalov *et al.*[2] observed a metal-insulator transition in silicon MOSFET inversion layers in the presence of weak disorder. This transition cannot be understood in terms of single-particle localization. For example, (i) the longitudinal resistance R_{xx} grows exponentially, (ii) the conductivity above and below the threshold does not fit with the variable range hopping model and (iii) capacitive measurements do not show a decrease in the effective conducting area. Their measurements on nonlinear dc transport, thermal activation energies for conductivity and threshold electric fields all indicate that the insulator state they observe is a some sort of collective state. They looked at pinned Wigner solids or charge density waves as possible candidates but could draw no definitive conclusions. The exact nature of the collective insulator state remains unclear.

Based on their experimental measurements Pudalov *et al.* presented a metal-insulator phase diagram as a function of the electron density n_s and the peak mobility μ^{peak}. The role of disorder in the sample can be characterized by μ^{peak}. To compare with their phase diagram we calculate μ^{peak} at the critical values of impurity concentration[20] n_{ci} using self-consistent mode-coupling theory.[21] The local field factor $G(\boldsymbol{q})$ used in Ref. 21 was approximated by a Hubbard expression for exchange only and this leads to higher peak mobilities than with our $G(\boldsymbol{q})$, which includes both exchange and correlations.

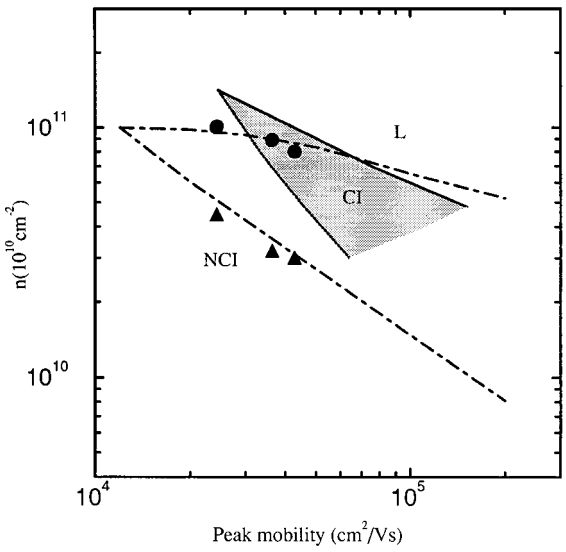

FIG. 2. Phase diagram as a function of electron density and peak mobility. Upper region is the conducting liquid phase L. The shaded region is our coherent insulator CI. Lower region is the noncoherent insulator. Experimental points and dot-dashed lines passing through them are from Ref. 2; circles, metal-insulator transition, triangles, transition from coherent localization to single-particle localization.

Figure 2 shows our calculated ergodic to nonergodic transition line. We also show the position of the metal-insulator transition (circles) for three silicon samples taken from Ref. 2. Their position is in good agreement with our line.

Pudalov *et al.* found with decreasing density or increasing disorder that the correlation length L_D (the domain size) decreases. This drives the coherent insulating state into a single-particle localization in which L_D becomes of the order of r_0. The crossover from coherent insulating state to single-particle localization is not precisely determined in the experiment. The criterion they decided upon for the crossover was that $L_D = 2r_0$. The three triangles mark these points in Fig. 2.

Near the transition our calculated electron-electron memory function $M_{ee}(\boldsymbol{q}) \gg M_{de}(\boldsymbol{q})$, the defect-electron memory function. If we decrease μ^{peak} while holding r_s fixed then $M_{de}(\boldsymbol{q})$ gets bigger. It is clear from Fig. 1(b) that $f(\boldsymbol{q})$ eventually evolves to a Gaussian-like function characteristic of single-particle localization. As in the experimental case the boundary between the two different types of localization is not precise and we take as our criterion for the cross-over that $M_{ee}(\boldsymbol{q}) \simeq M_{de}(\boldsymbol{q})$. The region bounded by the thick solid and dashed lines then depicts the phase region where the system is in the coherent localized state. The region below the dashed line is the noncoherent localized phase.

Pudalov *et al.* find when $r_s \lesssim 8$ that the metal-insulator transition goes directly to single-particle localization. This is again consistent with our results: for $r_s \lesssim 7$ we find only noncoherent localization. For noncoherent localization many-body effects are not dominant. The electrons localize independently, analogous to single-particle localization around impurities.

In summary the idealized (assumed zero hopping) classical glass transition method has been modified for the low-density electron system. The driving mechanism for the tran-

sition are the strong Coulomb correlations between electrons. We also include the effects of electron-impurity and interface surface-roughness scattering.

In the extreme low disorder limit our method reproduces the well-known intrinsic weak localization characteristic of low-dimensional systems. At small but finite disorder we get a transition to a coherent solid phase with liquidlike short-range order, which persists while electron-electron scattering dominates the dynamics but it gets destroyed at higher levels of disorder. This occurs when $M_{de}(\boldsymbol{q})$ starts competing with $M_{ee}(\boldsymbol{q})$. Similarly, the noncoherent localized phase replaces the coherent phase if we decrease the strength of the electron-electron interactions by increasing the electron density.

The agreement between our phase diagram and the recent experiments of Pudalov *et al.* strongly suggest that the coherent insulating phase observed in Ref. 2 is a *glassy phase*. Experimental evidence of electron glassy behavior has been reported in gallium arsenide samples of much less disorder than these silicon samples.[22] The possibility of a glassy coherent phase should be checked against other experiments. One possibility is a search for hysteresis which provides direct evidence for metastable states of a system.

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