Coherent Magnetotransport Through an Artificial Molecule

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The conductance in an extended multiband Hubbard model describing linear arrays of up to ten quantum dots is calculated via a Lanczos technique. A pronounced suppression of certain resonant conductance peaks in an applied magnetic field due to a density-dependent spin-polarization transition is predicted to be a clear signature of a coherent “molecular” wavefunction in the array. A many-body enhancement of localization is predicted to give rise to a giant magnetoconductance effect in systems with magnetic scattering.

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Arrays of coupled quantum dots can be thought of as systems of artificial atoms separated by tunable tunnel barriers. Two complementary theoretical approaches have been useful in describing such systems in the limit where charging effects are important but interdot tunneling is incoherent and in the limit of coherent ballistic transport, with charging effects neglected. However, recent improvements in fabrication and experimental techniques should make it possible to probe a third regime, where both interaction and coherence effects play nontrivial roles. In this regime, the system of coupled quantum dots behaves like an artificial molecule, and must be described by a coherent many-body wavefunction. In this Letter, we describe some striking characteristic signatures of such a coherent molecular wavefunction in the low-temperature magnetotransport through an array of quantum dots. Our theoretical predictions should be experimentally testable in currently available GaAs quantum dot systems.

An important consequence of coherent interdot tunneling is the formation of interdot spin-spin correlations analogous to those in a chemical bond at an energy scale \( \sim t^2/U \), where \( U = e^2/C_p \) is the charging energy of a quantum dot and \( t = (\hbar^2/2m^*) \int d^3x \Psi^*_j(x) \nabla^2 \Psi_i(x) \) is the interdot hopping matrix element, \( \Psi_j \) being electronic orbitals on nearest-neighbor dots. In an applied magnetic field, the Zeeman splitting drastically modifies the many-body wavefunction of the array when the Zeeman energy \( g \mu_B B \sim t^2/U \), and we find that certain resonant conductance peaks are suppressed (or, in some cases, enhanced) by several orders of magnitude compared to their size at \( B = 0 \). For quantum dots electrostatically defined in a 2D electron gas, no significant modification of the wavefunction of a single quantum dot would occur for magnetic fields of this magnitude in the plane of the dots (the case we consider), so that the standard Coulomb blockade-based transport theory neglects coherent interdot tunneling would predict no interesting magnetic field dependence of the conductance. Such spin-spin correlations are intrinsic many-body effects which are non-perturbative in the Coulomb interaction, and can not be explained by ballistic transport theories either. We therefore believe that observation of the predicted dramatic magnetic field effect on low-temperature transport through coupled quantum dots would represent the clearest possible signature of the formation of an “artificial molecule.”

The system we wish to model consists of a linear array of quantum dots electrostatically defined in a 2D electron gas, with a magnetic field in the plane of the dots. We neglect intradot correlations (a reasonable approximation if the number of electrons per dot is not too small) and focus instead on collective phenomena in the array. The electron-electron interactions in the array are described by a capacitance matrix \( C_{ij} \): we assume constant capacitances \( C_g \) between each quantum dot and the macroscopic metallic gate which defines its confinement potential, and a capacitance \( C_l \), which is a function of gate voltage and may include important quantum mechanical corrections, between nearest neighbor quantum dots. The electronic orbitals in the confining potential of an isolated dot are taken to be nondegenerate with level spacing \( \Delta \), and the hopping matrix element \( t_{ij} \) between (nearly) degenerate orbitals on nearest-neighbor dots is assumed largest, all others being neglected. The Hamiltonian is

\[
\hat{H} = \sum_{j, n, \sigma} \left( t_n \hat{c}^\dagger_{j+1n\sigma} \hat{c}_{jn\sigma} + \text{H.c.} \right) + \sum_{j, n, \sigma} \left( n \Delta + \sigma g \mu_B B/2 \right) \hat{c}^\dagger_{jn\sigma} \hat{c}_{jn\sigma} + \frac{e^2}{2} \sum_{i, j} C_{ij}^{-1} \hat{n}_i \hat{n}_j, \tag{1}
\]

where \( \hat{c}^\dagger_{jn\sigma} \) is the creation operator for an electron of spin \( \sigma \) in the \( n \)th orbital of the \( j \)th dot, \( \hat{n}_j = \sum_{n, \sigma} \hat{c}^\dagger_{jn\sigma} \hat{c}_{jn\sigma} \), and the sums run from \( n = 0 \) to \( M - 1 \) (the \( M \) orbitals nearest the Fermi energy \( E_F \)). In the strongly-correlated regime, the interaction term in Eq. (1) cannot be treated perturbatively. We therefore employ a Lanczos technique to compute the exact many body ground states of arrays of 5 to 12 quantum dots with 1 to 5 electronic orbitals per dot.
The array is coupled to noninteracting leads via a tunneling Hamiltonian with matrix elements \( t_{n,\sigma}^{r,l} \times t_{n} \) which couple electrons in the \( n \)th orbital of the 1st \((L)\) dot to the right (left) lead. The capacitance to the leads is neglected. In the limit \( \Delta E \gg k_B T \gg \hbar \Gamma \), where \( \Delta E \) is the energy level spacing in the array and \( \Gamma \) is the tunneling rate of electrons out of the array, the linear response conductance is determined by ground state to ground state transitions, and is given by

\[
G = e^2 \sum_{N} \frac{\Gamma_N^{r} \Gamma_N^{l}}{\Gamma_N + \Gamma_N^{r,l}} A_N(\mu),
\]

where \( \Gamma_N^{r,l} = 2\pi \sum_{n,\sigma} |\langle 0_N | t_{n}^{r,l} c_{i(L)}^{\dagger} n_{\sigma} | 0_N-1 \rangle|^2 \rho_{n}^{r,l}(E_N^0 - E_N^{0,-1})/\hbar \), \( \rho_{n}^{r,l}(\varepsilon) \) being the density of states in the leads.

For the case \( B > 0 \), the ground state is non-degenerate and \( A_N(\mu) = -f'(E_N^0 - E_N^{0,-1}) \), while for \( B = 0 \), the ground state is spin-degenerate when \( N \) is odd and \( A_N(\mu) = 2/[k_B T(3 + 2e^{-\varepsilon_N} + e^{\varepsilon_N})] \), where \( x_N = (-1)^N (\mu - E_N^0 - E_N^{0,-1})/k_B T \). Eq. 2 is derived by the method of Refs. 6, 8.

Fig. 1 shows the conductance through a linear array of 10 quantum dots with \( C_i = 0 \) as a function of the chemical potential \( \mu \) in the leads, whose value relative to the energy of the array is controlled by the gate voltages. The two Coulomb blockade peaks in Fig. 1 are split into multiplets of 10 by interdot tunneling, as discussed in Refs. 5, 6. We refer to these multiplets as Hubbard minibands. The energy gap between multiplets is caused by collective Coulomb blockade [4], and is analogous to the energy gap in a Mott insulator [5]. The heights of the resonant conductance peaks in Fig. 1(a) can be understood as follows: Since the barriers to the leads are assumed to be large, the single-particle wavefunctions of the array are like those of a particle in a one-dimensional box. The lowest eigenstate has a maximum in the center of the array and a long wavelength, hence a small amplitude on the end dots, leading to a suppression of the 1st conductance peak. Higher energy single-particle states have shorter wavelengths, and hence larger amplitudes on the end dots, leading to conductance peaks of increasing height. The suppression of the conductance peaks at the top of the 1st miniband can be understood by an analogous argument in terms of many-body eigenstates; the 10th electron which enters the array can be thought of as filling a single hole in a Mott insulator, etc.

In Fig. 1(b), the spin-degeneracy of the quantum dot orbitals is lifted by the Zeeman splitting. There is a critical field \( B_c \) above which the system is spin-polarized; for an infinite 1D array with \( C_i = 0 \) and \( t \ll U \),

\[
g \mu B_c \simeq \frac{4t^2}{\pi U}(2\pi n - \sin 2\pi n),
\]

where \( n < 1 \) is the filling factor of the lower Hubbard band. (Recall that we are here considering only the single spin-1/2 orbital nearest \( E_F \) in each quantum dot—the magnetic field required to spin-polarize an entire quantum dot is much larger.) Because \( B_c \) is a function of \( n \), one can pass through this spin-polarization transition (SPT) by varying \( n \) at fixed \( B \). In Fig. 1(b), this transition occurs between the 4th and 5th electrons added to the array, consistent with the prediction of Eq. (3). The effect of this transition on the conductance spectrum is dramatic: The first 4 electrons which enter the array have spin aligned with \( B \) (up), but the 5th electron enters with the opposite spin, and goes predominantly into the lowest single-particle eigenstate for down-spin electrons, which couples only weakly to the leads, leading to a suppression of the 5th resonant conductance peak by over an order of magnitude. It should be emphasized that the heights of the conductance peaks change discontinuously as a function of \( B \) each time there is a spin-flip.

Splitting of the Coulomb blockade peaks due to interdot coupling and suppression of the conductance peaks at the miniband edges have recently been observed experimentally by Waugh et al. [2]. However, it has been pointed out [2] that both effects can also be accounted for by a model [4] of capacitively coupled dots with completely incoherent interdot tunneling. It is therefore of interest to consider the effects of interdot capacitative coupling in the regime of coherent interdot transport. A nonzero interdot capacitance \( C_i \) introduces long-range electron-electron interactions in Eq. (1) and decreases the intradot charging energy \( U \). Fig. 2 shows the spin susceptibility \( \chi_s \) for \( C_i/C_B = 1/2 \) in linear arrays with 8 electrons on 12 dots and 10 electrons on 10 dots. The \( n \)-dependence of \( B_c \) in Fig. 3 is qualitatively similar to that in a system with intradot interactions only, but the values of \( B_c \) are roughly twice those of a system with \( C_i = 0 \). Note the rapid growth of \( \chi_s \) as \( B \rightarrow B_c \).
FIG. 2. Spin susceptibility $\chi_s = \hbar^{-1} \Delta S/\Delta B$ at $T = 0$ vs. magnetic field $B$ for linear arrays of GaAs quantum dots with $e^2/C_g = 1\text{meV}$, $C_i/C_g = 0.5$, and $t = 0.05\text{meV}$. Squares: 10 electrons on 10 dots ($B_c \approx 1.5T$); triangles: 8 electrons on 12 dots ($B_c \approx 1T$).

an infinite array, $\chi_s$ is expected to diverge as $B \to B_c$ because the system undergoes a second order quantum phase transition [13]. The SPT predicted to occur in an array of coupled quantum dots is in contrast to that observed in a single quantum dot [14], where the critical point occurs for minimum total spin.

Disorder introduces a length scale which cuts off the critical behavior as $B \to B_c$. However, as shown in Fig. 3, where disorder $\delta t/\bar{t} \sim 1$ ($t_{i\uparrow} \neq t_{i\downarrow}$) has been included in the hopping matrix elements, the SPT has a clear signature in the magnetotransport even in a strongly disordered system. In Fig. 3, the peak splitting due to capacitive coupling is roughly ten times that due to interdot tunneling, so that the peak positions are within $\sim 10\%$ of those predicted by a classical charging model [3]. However, the dramatic dependence of peak heights on magnetic field—the 4th conductance peak in Fig. 3(b) is suppressed by a factor of 32 compared to its $B = 0$ value due to the density-dependent SPT described above—can not be accounted for in a model which neglects coherent interdot tunneling. This effect should be observable provided $g\mu_B B_c > \max(k_B T, \hbar/\tau_i)$, where $\tau_i$ is the inelastic scattering time. We believe that this striking magnetotransport effect is the clearest possible signature of a coherent molecular wavefunction in an array of quantum dots.

Fig. 4 shows the conductance spectrum for an array of 6 quantum dots with the same parameters as in Fig. 3, but with spin-dependent disorder in the hopping matrix elements, as could be introduced by magnetic impurities. Several conductance peaks at $B = 0$ (solid curve) are strongly suppressed due to a many-body enhancement of localization. This effect arises because repulsive on-site interactions enhance spin-density wave correlations, which are pinned by the spin-dependent disorder [15]. At $B = 1.3T$ (dotted curve) the system is above $B_c$ and is spin-polarized, circumventing this effect. The second conductance peak is enhanced by a factor of 1600 at 1.3T compared to its size at $B = 0$ (not visible on this scale). This giant magnetoconductance effect is a many-body effect intrinsic to the regime of coherent interdot transport.

Another interesting phenomenon stemming from the competition between coherent interdot tunneling and charging effects is the Mott-Hubbard metal-insulator transition (MH-MIT), which occurs when collective Coulomb blockade (CCB) [5] is destroyed due to strong interdot coupling. For GaAs quantum dots larger than about 100nm in diameter, we find that this transition is caused by the divergence of the effective interdot
capacitance, similar to the breakdown of Coulomb blockade in a single quantum dot. Within the framework of the scaling theory of the MH-MIT, one expects a crossover from CCB to ballistic transport in a finite array of quantum dots when the correlation length $\xi$ in the CCB phase significantly exceeds the linear dimension $L$ of the array. Fig. 3 shows the conductance spectrum for 5 quantum dots with 5 spin-1/2 orbitals per dot. The divergence of the effective interdot capacitance as the interdot barriers become transparent is simulated by setting $C_i^{(n)}/C_g = 2^n$, $n = 0, \ldots, 4$. In Fig. 3, minibands arising from each orbital are split symmetrically into multiplets of 5 peaks by CCB, with the center to center spacing between multiplets equal to $e^2/C_g$, while the energy gap between minibands corresponds to the band gap $\Delta$ enhanced by charging effects. The CCB energy gap is evident in the first 3 minibands, but is not resolved for the higher orbitals ($C_i/C_g \geq 4$), although there is still a slight suppression of the conductance peaks near the center of the 4th miniband. Comparison of the compressibility of the system to a universal scaling function for the MH-MIT calculated by the method of Ref. 12 indicates that the MH-MIT probably occurs at $C_i/C_g = \infty$ in an infinite array of quantum dots, when the interdot barriers become transparent to one transmission mode.

In conclusion, we predict that low-temperature magnetotransport experiments on judiciously fabricated quantum dot arrays would lead to the observation of a variety of phenomena resulting from the interplay between coherent interdot tunneling and charging effects: SPT, MH-MIT, and giant magnetoconductance due to a many-body enhancement of localization by magnetic scattering.

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