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Coherent magnetotransport through an artificial molecule

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Abstract

Coherent resonant tunneling through a double quantum dot in an inhomogeneous magnetic field is investigated using a generalized Hubbard model. The conductance is calculated analytically for the case of a single spin-1/2 orbital per dot and numerically for dots containing multiple orbitals using a multi-terminal Breit–Wigner type formula, which allows for the explicit inclusion of inelastic processes. Giant spin-dependent many-body corrections to the transport are predicted to be a clear signature of the formation of a molecular-like state in the system. © 1997 Elsevier Science B.V.

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Arrays of coupled quantum dots [1–6] 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 [7] where charging effects are important but interdot tunneling is *incoherent* and in the limit [8] of coherent *ballistic* transport, with charging effects neglected. However, recent improvements in fabrication and experimental techniques now make it possible to probe a third regime, where both interaction *and* coherence effects play nontrivial roles [3–6]. In this regime, the system of coupled quantum dots behaves like an artificial molecule, and must be described by a coherent many-body wavefunction [9–11]. In this Letter, we describe some striking characteristic signatures of such a coherent molecular wavefunction in the low-temperature magnetotransport through a double quantum dot. 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 [12] analogous to those in a chemical bond at an energy scale $J \sim t^2/U$, where U is the charging energy of a quantum dot and $t = (\hbar^2/2m^*) \int d^3x\, \Psi_m^*(x) \nabla^2 \Psi_n(x)$ is the interdot hopping matrix element, $\Psi_{m,n}$ being electronic orbitals on nearest-neighbor dots. In a system with magnetic disorder, such a spin configuration is pinned, and the resulting blockage of spin backflow [13] leads to strong charge localization. However, an applied magnetic field will break such an antiferromagnetic bond when the Zeeman splitting $g\mu_B B > J$, leading to an enormous enhancement of the charge mobility. Such spin-dependent many-body effects on the magnetotransport should be experimentally observable provided $\Gamma + \Gamma^{(i)}, k_B T \lesssim J$, where $\Gamma + \Gamma^{(i)}$ is the total broadening of the resonant levels of the system; they can be readily distinguished from orbital effects in arrays of quasi-two-dimensional quantum dots by applying the magnetic field *in the plane* of the dots. Observation

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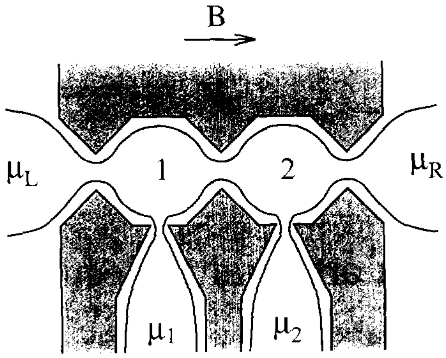

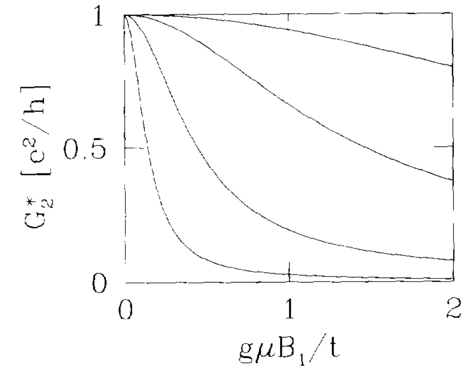

Fig. 1. Schematic diagram of a double quantum dot.

Fig. 2. Resonant conductance at $T = 0$ in units of $(e^2/h)/(1+\Gamma^{(i)}/\Gamma)$ as a function of the Zeeman splitting on dot 1 for $\gamma^{-1} = 0, 3, 10, 30$ (top to bottom). If an additional uniform external field $B > J$ is applied, the conductance is restored to the value for $\gamma^{-1} = 0$, leading to giant magnetoresistance.

of the predicted giant magnetoresistance effect in the low-temperature transport through coupled quantum dots would, we believe, represent a clear signature of the formation of an artificial molecular bond.

The system under consideration (Fig. 1) consists of a double quantum dot electrostatically defined [2–6] in a 2D electron gas, coupled weakly to several macroscopic electron reservoirs, with a magnetic field in the plane of the dots. Each quantum dot is modeled by 1–4 spin-1/2 orbitals, representing the electronic states nearest the Fermi energy E_F, and is coupled via tunneling to its neighbor and to one or more electron reservoirs. Transport occurs between the left (L) and right (R) reservoirs; reservoirs 1 and 2 are considered to be ideal voltage probes [15], and serve to introduce inelastic processes in the system [16]. Electron–electron interactions in the array are described [7,14] by a matrix of capacitances C_{ij}: we assume a capacitance C_g between each quantum dot and the system of metallic gates held at voltage V_g, an interdot capacitance C_i, and a capacitance C_r between a quantum dot and each of its associated electron reservoirs. The diagonal elements of C_{ij} are the sum of all capacitances associated with a quantum dot, $C_\Sigma = C_g + C_i + 2C_r$, and the off-diagonal elements are $-C_i$. These capacitance coefficients may differ from their geometrical values due to quantum mechanical corrections [11,17], but enter only as parameters in our model. The Hamiltonian of the system is

$$H = H_{\rm dots} + \sum_{\sigma,\alpha}\sum_{k\in\alpha} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{\sigma,\alpha,j}\sum_{k\in\alpha}\sum_{m\in j}(V_{km} c_{k\sigma}^\dagger d_{m\sigma} + \text{H.c.}), \tag{1}$$

where $d_{m\sigma}^\dagger$ $(m \in j)$ creates an electron of spin σ in orbital m of the jth dot and $c_{k\sigma}^\dagger$ $(k \in \alpha)$ creates an electron in state $k\sigma$ of reservoir α. Here

$$H_{\rm dots} = \sum_{\sigma,j}\sum_{m\in j} \epsilon_{m\sigma} d_{m\sigma}^\dagger d_{m\sigma} + \sum_{\sigma}\sum_{m\in 1}\sum_{n\in 2}(t_{mn} d_{m\sigma}^\dagger d_{n\sigma} + \text{H.c.}) + \tfrac{1}{2}\sum_{i,j}(Q_i + Q_g) C_{ij}^{-1}(Q_j + Q_g), \tag{2}$$

where $Q_j \equiv -e\sum_{\sigma,m\in j} d_{m\sigma}^\dagger d_{m\sigma}$, $Q_g \equiv C_g V_g$, and $\epsilon_{m\sigma}\,(m \in j) = \epsilon_m + \sigma B_j/2$, where B_j is the Zeeman splitting on dot j and ϵ_m is the energy of the mth quantum-confined orbital on dot j. For the case of a single orbital per dot, $H_{\rm dots}$ reduces to a two-site Hubbard model with on-site repulsion $U = e^2 C_\Sigma/(C_\Sigma^2 - C_i^2)$ and nearest neighbor repulsion $V = e^2 C_i/(C_\Sigma^2 - C_i^2)$. A similar model for a double quantum dot has recently been studied for the case of a large number of orbitals per dot with $B_j = 0$ by Matveev, Glazman, and Baranger, and by Golden and Halperin [11].

The N-body ground states $|0_N\rangle$ of the isolated double dot are nondegenerate, with energy E_N^0, except at a finite set of magnetic field values $\{B_N^m,\ m = 1,\dots,\text{Int}(N/2)\}$, where spin-flip occurs. At these special fields, the ground state is doubly degenerate and the low-temperature physics of the system is that of the Kondo effect, discussed in Ref. [18]. For general B, however, the current through the system will be determined by transitions $|0_{N-1}\rangle \rightleftarrows |0_N\rangle$ between nondegenerate ground states, provided the tunnel barriers to the reservoirs are sufficiently large, and provided the temperature and bias are small compared to the energy of an excitation. In this limit, the system can be shown to exhibit Breit-Wigner type resonances, and the expectation value of the current flowing into reservoir α can be expressed using the multiprobe current formula [19]

$$I_\alpha = \frac{e}{h}\sum_{\beta=1}^{M}\int d\epsilon \sum_N \frac{\Gamma_N^\alpha \Gamma_N^\beta [f_\alpha(\epsilon) - f_\beta(\epsilon)]}{(\epsilon - E_N^0 + E_{N-1}^0)^2 + (\Gamma_N/2)^2}, \tag{3}$$

where $f_\alpha(\epsilon) = \{\exp[(\epsilon - \mu_\alpha)/k_B T] + 1\}^{-1}$ is the Fermi function for reservoir α,

$$\Gamma_N^\alpha = 2\pi \sum_{k\in\alpha}\sum_{n,m,\sigma} \langle 0_{N-1}|V_{kn} d_{n\sigma}|0_N\rangle \langle 0_N|V_{km}^* d_{m\sigma}^\dagger|0_{N-1}\rangle \delta(\epsilon_k - E_N^0 + E_{N-1}^0), \tag{4}$$

and $\Gamma_N = \sum_\alpha \Gamma_N^\alpha$. We demand that the expectation value of the current flowing into reservoirs 1 and 2 (which serve as ideal voltage probes) be zero, which fixes $\mu_{1,2}$ via Eq. (3). Eliminating $f_{1,2}(\epsilon)$ from Eq. (3) and taking the linear response limit, one finds the two-terminal conductance

$$G = \frac{e^2}{h}\sum_N \frac{\Gamma_N^L \Gamma_N^R}{\Gamma_N^L + \Gamma_N^R}\int \frac{\Gamma_N[-f'(\epsilon)]\, d\epsilon}{(\epsilon - E_N^0 + E_{N-1}^0)^2 + (\Gamma_N/2)^2}. \tag{5}$$

The total width of the Nth resonance may be written $\Gamma_N = \Gamma_N^L + \Gamma_N^R + \Gamma_N^{(i)}$, where the quantity $\Gamma_N^{(i)}/\hbar = (\Gamma_N^1 + \Gamma_N^2)/\hbar$ may be interpreted as the total inelastic scattering rate due to phase-breaking processes in the auxiliary reservoirs [16]. It should be emphasized that both the elastic and inelastic broadening of the conductance resonances are suppressed by the many-body factors in Eq. (4), which lead to an orthogonality catastrophe [20] in the large-N limit. The effect of such an orthogonality catastrophe in the sequential tunneling regime has previously been discussed by Kinaret et al. [21] and by Matveev, Glazman, and Baranger [11].

Let us first consider the case of a single spin-1/2 orbital per dot, for which the conductance matrix elements (4) can be obtained analytically. For simplicity, we assume that the tunnel barriers coupling the system to the external reservoirs are described by the energy-independent parameters $2\pi\sum_{k\in\alpha}|V_\alpha|^2\delta(\epsilon_k - E) = \Gamma$, $\alpha = L, R$; $\Gamma^{(i)}$, $\alpha = 1, 2$. Then the conductance matrix elements depend only on the many-body wavefunctions of the double dot. Spin disorder is introduced via a Zeeman splitting on dot 1, $B_1 = 4t\Delta$, $B_2 = 0$. Experimentally, such an inhomogeneous field could be produced, e.g., by the presence of a small ferromagnetic particle. The total width of the one-particle resonance is found to be $\Gamma_1 = \Gamma + \Gamma^{(i)}$, and the prefactor in Eq. (5) is $\Gamma_1^L\Gamma_1^R/(\Gamma_1^L + \Gamma_1^R) = (\Gamma/4)/(1+\Delta^2) \equiv \Gamma_0$. The maximum conductance at the one-particle resonance is thus

$$\begin{aligned} G_1^* &= \frac{e^2/h}{(1+\Delta^2)(1+\Gamma^{(i)}/\Gamma)}, & T &= 0, \\ &= e^2\Gamma_0/4\hbar k_B T, & \Gamma + \Gamma^{(i)} &\ll k_B T \ll t\sqrt{1+\Delta^2}. \end{aligned} \tag{6}$$

Inelastic scattering suppresses the resonant conductance at $T = 0$, but has no effect when the resonance is thermally broadened. For $U - V \gg t$, the two-particle ground state of the double quantum dot has an antiferromagnetic spin configuration characterized by the superexchange parameter

$$J = 2t(\gamma - \Delta + \sqrt{\gamma^2 + \Delta^2}), \tag{7}$$

where $\gamma = t/(U - V)$. Note that $2t\gamma \leqslant J \leqslant 4t\gamma$. The two-particle resonance is separated from the one-particle resonance by $e\Delta Q_g/(C_\Sigma - C_i) = V + 2t(1+\Delta^2)^{1/2} - J$, and the conductance is determined by the matrix elements

$$\langle 0_2|d^\dagger_{j\uparrow}|0_1\rangle = \frac{\sqrt{2}}{A}\left(\frac{2\gamma}{[1+(\Delta\mp\sqrt{\Delta^2+1})^2]^{1/2}} + \frac{1\mp\Delta/(\gamma+\sqrt{\gamma^2+\Delta^2})}{[1+(\Delta\pm\sqrt{\Delta^2+1})^2]^{1/2}}\right), \tag{8}$$

where $A^2 = 1 + \Delta^2/(\gamma+\sqrt{\gamma^2+\Delta^2})^2$ and the upper (lower) sign holds for $j = 1$ (2). For $B_1 \gtrsim J$, the antiferromagnetic spin configuration is pinned, leading to a strong suppression of the amplitude to inject electron 2 into dot 1, and a concommitant suppression of the second conductance peak. Inserting Eq. (8) into Eqs. (4) and (5), one finds the $T = 0$ resonant conductance

$$G_2^* = \frac{16(e^2/h)(\gamma/\Delta)^2}{1+\Gamma^{(i)}/\Gamma}, \qquad \gamma \ll \Delta \ll 1,$$
$$= \frac{4(e^2/h)\gamma^2}{1+\Gamma^{(i)}/\Gamma}, \qquad \Delta \gg 1. \tag{9}$$

A second doublet of conductance peaks for $N = 3, 4$ is separated from this doublet by $\Delta Q_g \simeq e$ (center to center), and one finds $G_3^* = G_2^*$, $G_4^* = G_1^*$ due to electron-hole symmetry. The resonant conductance for $N = 2$ is suppressed by a factor of γ^2 compared to that for $N = 1$ due to collective spin pinning (one readily verifies that the resonant conductance is suppressed by the same many-body factor in the regime of thermally broadened resonances). This dramatic many-body suppression of the conductance is illustrated in Fig. 2 for several values of γ. The effect of spin disorder is to be contrasted with that of a charge detuning $\Delta = (\epsilon_1 - \epsilon_2)/2t$, investigated by Klimeck et al. [10] and by van der Vaart et al. [3], for which both G_1^* and G_2^* are given by Eq. (6) at $T = 0$ (G_2^* is then reduced by a factor of 2 in the thermally broadened regime). The very different effects of spin and charge disorder stem from the fact that the repulsive interactions in Eq. (2) enhance spin-density fluctuations, but suppress charge-density fluctuations.

Let us now consider the effect of an additional homogeneous magnetic field applied parallel to the inhomogeneous field, $B_1 = 4t\Delta + B$, $B_2 = B$. For $B > J$, it is energetically favorable to break the antiferromagnetic bond between the dots and form a spin-polarized state, thus preventing collective spin pinning effects. G_2^* is then given by Eq. (6). The resulting magnetoresistance on resonance for $T = 0$ and $\Delta \gg 1$ is thus

$$\frac{\Delta R^*}{\Delta B} = -\frac{h}{e^2}\frac{g\mu_B}{4J\gamma^2}(1+\Gamma^{(i)}/\Gamma) \sim -\frac{h}{e^2}\frac{g\mu_B(U-V)^3}{8t^4}. \tag{10}$$

In the thermally broadened resonance regime, the factor $1 + \Gamma^{(i)}/\Gamma$ is replaced by $2k_BT/\pi\Gamma$. Since the Coulomb energy $U - V$ is typically large compared to the interdot tunneling matrix element t, the predicted magnetoresistance is extremely large. This giant magnetoresistance effect is a direct indication of the field-induced breaking of the artificial molecular bond between the dots [1].

The conditions necessary to observe the predicted magnetoresistance effect may be determined by including the effect of transport through the triplet excited state via the method of Refs. [10,14]. One finds the resonant conductance at $B = 0$ for $k_BT \gg \Gamma + \Gamma^{(i)}$,

$$G_2^* = \frac{e^2}{2\hbar k_BT}\frac{\exp(\beta J)}{2\exp(\beta J)-1}\left(\Gamma_s + \frac{2\Gamma_t}{\exp(\beta J)+1}\right), \tag{11}$$

where $\Gamma_s \simeq 4\gamma^2\Gamma$ is the sequential tunneling rate through the pinned antiferromagnetic ground state and $\Gamma_t = \Gamma_0$ is the sequential tunneling rate through the triplet excited state. The magnetoresistance is thus reduced by a factor of 2 at a temperature $k_BT_{1/2} = J/\ln(\Gamma_t/\Gamma_s)$. Increased coupling to the leads and/or inelastic scattering can be shown [22] to lead to a similar admixture of transport through excited states when $\Gamma + \Gamma^{(i)} \sim J$. We therefore expect the predicted giant magnetoresistance effect to be observable for k_BT, $\Gamma + \Gamma^{(i)} \lesssim J$. In

[1] The parallel quantum dot geometry of Hofmann et al. [5] should exhibit a similar magnetoresistance effect in the thermally broadened resonance regime.

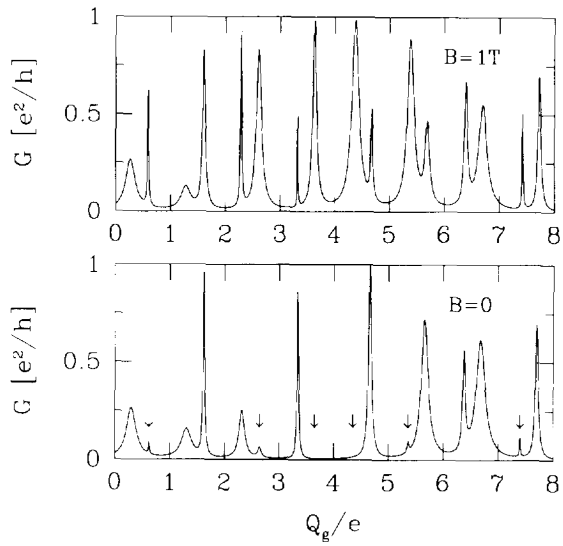

Fig. 3. Zero temperature conductance of a double quantum dot with inhomogeneous field $B_1 - B_2 = 1.3$ T as a function of the gate voltage $Q_g = C_g V_g$. Each dot has four spin-1/2 orbitals with random energies and mean level spacing 0.03 meV. The orbitals on neighboring dots are coupled by random tunneling matrix elements with mean $\bar{t} = .015$ meV. The coupling of the dot orbitals to the external reservoirs is random with mean value $\bar{\Gamma} = 4$ μeV ($\Gamma^{(i)} = 0$). $e^2/C_\Sigma = 1$ meV and $C_i/C_\Sigma = 1/3$. Conductance peaks suppressed at $B = 0$ due to collective spin pinning are marked by arrows.

currently available GaAs quantum dot systems, charging energies are typically of order 1 meV, and one expects tunneling matrix elements $t \sim 0.1$ meV for moderate to strong interdot tunneling, so values of J in the range 0.01–0.1 meV should be attainable.

The binding energy J of the artificial molecular bond formed in coupled quantum dots is enhanced for strong interdot tunneling. However, for sufficiently strong interdot tunneling, the single-orbital approximation employed in the analytical calculation above may break down, as discussed in Refs. [4,11]. Let us therefore consider a double quantum dot with four spin-1/2 orbitals per dot, described by Eqs. (1) and (2) with random ϵ_m, t_{mn}, and V_{km}, as specified in Fig. 3. For these parameters, the bandwidth of the interdot tunneling is comparable to the width of the level distribution in each dot. A Zeeman term corresponding to a local field of 1.3 T is included on dot 1. Eq. (2) is solved numerically via the Lanczos technique, and the conductance calculated at $T = 0$ using Eq. (5) is shown as a function of gate voltage in Fig. 3. When an additional homogeneous field $B = 1$ T exceeding the estimated interdot spin–spin interaction is applied to the system (upper curve), the conductance exhibits a sequence of eight doublets, corresponding to the successive addition of 16 electrons to the double dot. The splitting within a doublet is due to interdot tunneling and Coulomb interactions, as discussed above and in Refs. [4,10,11], while the splitting between doublets arises due to collective Coulomb blockade [9]. Similar conductance spectra have recently been observed experimentally in double quantum dots by Waugh et al. [4] and by Blick et al. [6]. At $B = 0$, however, several conductance peaks are strongly suppressed, though both peaks within a doublet are never suppressed simultaneously (a similar pattern of peak suppression was obtained for several other realizations of disorder). The magnitude of the conductance peak suppression varies from doublet to doublet due to the random interdot coupling t_{mn}, which implies a random superexchange J, but is consistent with Eq. (9) for large Δ. The predicted giant magnetoresistance effect thus persists even when the bandwidth of interdot tunneling exceeds the level spacing of a quantum dot.

In conclusion, we have shown that the formation of an artificial molecular bond due to interdot superexchange can drastically modify the low-temperature transport through coupled quantum dots in an inhomogeneous magnetic field. The giant magnetoresistance effect proposed here for coupled quantum dots is expected to be quite general in narrow-band strongly correlated systems with magnetic disorder.

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