PHYSICAL REVIEW B **67**, 161307(R) (2003)

Nonlinear cotunneling through an artificial molecule

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(Received 3 December 2002; revised manuscript received 21 January 2003; published 30 April 2003)

We study electron transport through a system of two lateral quantum dots coupled in series. We consider the case of weak coupling to the leads and a bias point in the Coulomb blockade. After a generalized Schrieffer-Wolff transformation, cotunneling through this system is described using methods from lowest-order perturbation theory. We study the system for arbitrary bias voltages below the Coulomb energy. We observe a rich, non-monotonic behavior of the stationary current depending on the internal degrees of freedom. In particular, it turns out that at fixed transport voltage, the current through the system is largest at weak-to-intermediate interdot coupling.

DOI: 10.1103/PhysRevB.67.161307 PACS number(s): 73.63.Kv, 73.23.Hk, 72.10.−d, 03.67.Lx

Quantum dots are prototype systems for studying the properties of discrete levels embedded in a solid-state environment. Single dots (''artificial atoms''[1]) can be coupled through quantum point contacts, leading to ''artificial molecules.'' Indeed it has been shown experimentally[2–4] that the eigenstates of double-dot systems are coherent molecular superpositions of single dot (atomic) states. Unlike real molecules, these dots are readily contacted and tunable *in situ*, making them a natural test bed for molecular transport. Double dots have also been proposed as charge quantum bits.[5,6]

This raises the question, which information on the energy spectrum and the wave functions of the dot can be probed by transport measurements. This is only possible if artifacts induced by the coupling to the leads can be sorted out and the double-dot is disturbed as little as possible. This is the case when the coupling to the outside leads is weak (see Fig. 1) and the gates are tuned to the Coulomb blockade regime.[7,8] In that regime, only states with a fixed number of electrons are energetically permissible and hence sequential tunneling is suppressed. The leading transport mechanism in this case is cotunneling,[11] the coherent transfer of two electrons via virtual levels in the dots. Our work stands between studies focusing on sequential tunneling[9] and work on *linear response* in the Kondo regime.[10] The properties of cotunneling currents as a spectroscopic tool for the spectrum of quantum dot system have recently been studied in exquisitely controlled experiments on systems similar to ours.[4,12]

In this paper, we analyze a serial configuration of lateral quantum dots in the cotunneling regime. We study finite voltages up to the order of the charging energy, i.e., do not restrict ourselves to linear response. We find a rich nonmonotonic structure in the current as a function of the dot parameters. In particular, we find a pronounced crossover indicating the opening of an inelastic transport channel, which leads to the surprising result, that a too strong interdot coupling actually inhibits charge transport. We analyze the influence of the asymmetry of the dots on the current.

In the Coulomb blockade regime,[7,8] the relevant Hilbert space is spanned by four basis states $|i,j\rangle$, $i,j\in\{0,1\}$, which denotes i and j additional electrons (as compared to an appropriate neutral state) on the left and right dots, respectively. We study the situation where the gate voltages of the single dots are very close to each other and the interdot coupling is, although appreciable, much smaller than the single-dot addition energy. Thus, the subspace spanned by the two states $|1,0\rangle$ and $|0,1\rangle$ is energetically most favorable. The next closest states $|v_0\rangle=|0,0\rangle$ and $|v_2\rangle=|1,1\rangle$ are outside the transport window and serve as virtual states.[11] States with higher dipolar moment are energetically even less favorable due to the high charging energy of the individual dots.

The Hamiltonian of this system can be written as

$$H=H_0+H_1, \tag{1}$$

$$H_0=H_{\rm sys}+H_{\rm res}, \tag{2}$$

$$H_{\rm sys}=\varepsilon_{\rm as}(\hat{n}_l-\hat{n}_r)-\varepsilon_\alpha\hat{n}_{v_0}+\varepsilon_\beta\hat{n}_{v_2}+\gamma\sum_n\,(a_n^{L\dagger}a_n^R+a_n^{R\dagger}a_n^L), \tag{3}$$

$$H_{\rm res}=\sum_{\vec{k}}\,\varepsilon_{\vec{k}}^L b_{\vec{k}}^{L\dagger}b_{\vec{k}}^L+\sum_{\vec{k}'}\,\varepsilon_{\vec{k}'}^R b_{\vec{k}'}^{R\dagger}b_{\vec{k}'}^R, \tag{4}$$

$$H_1=t_c\sum_{\vec{k},n}\,(a_n^{L\dagger}b_{\vec{k}}^L+a_n^L b_{\vec{k}}^{L\dagger})+t_c\sum_{\vec{k}',n}\,(a_n^{R\dagger}b_{\vec{k}'}^R+a_n^R b_{\vec{k}'}^{R\dagger}). \tag{5}$$

Note that the sum over dot states n only runs over the restricted Hilbert space described above. H_0 describes the isolated double-dot ($H_{\rm sys}$) and the leads ($H_{\rm res}$), whereas the tunneling part H_1 describes the coupling of each dot to its

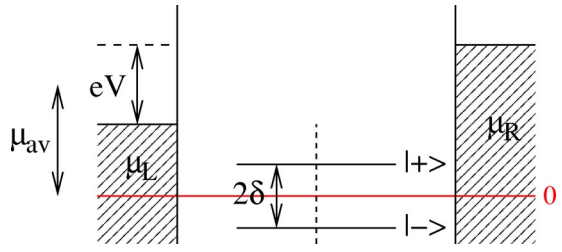

FIG. 1. Sketch of the considered artificial molecule, where 2δ is the level splitting and V is the bias voltage. The coupling to the outside leads (hatched areas) is assumed to be small whereas the interdot coupling (dotted line) can be strong.

lead and will be treated as a perturbation. $\hat{n}_{l/r}$ are the number operators counting additional electrons on either dot. The asymmetry energy $\varepsilon_{\rm as}=(\varepsilon_l-\varepsilon_r)/2$ describes half of the difference between the energy level for the additional electron in left dot (ε_l) and the corresponding energy level in the right dot (ε_r), which can be tuned through the gate voltages. ε_β and ε_α are the charging energies towards the higher level $|v_2\rangle$ and the lower level $|v_0\rangle$, respectively. γ is the tunable interdot coupling strength. The $a^{(\dagger)}$s and $b^{(\dagger)}$s denote electron creation/annihilation operators in the dots and leads. In H_1, the symbol t_c represents the tunnel matrix element between the dots and the leads. It is independent of the energies in the double-dot system and the corresponding sequential tunneling rate $\hbar\Gamma=2\pi t_c^2 N(\varepsilon_{\rm F})$ should be small compared to the internal energies. $N(\varepsilon_{\rm F})$ is the density of states in the leads taken at the Fermi energy. We restrict our analysis to spin-polarized electrons, these can be polarized by an appropriate in-plane magnetic field. Figure 1 shows a sketch of the system. In Fig. 1, $V=\mu_R-\mu_L$ is the bias voltage between the two leads (hatched) and $2\delta=2\sqrt{\varepsilon_{\rm as}^2+\gamma^2}$ is the level splitting in the molecular two-state system.

Pursuing our aforementioned objective, we take the interdot coupling γ into account to all orders by diagonalizing $H_{\rm sys}$ and transforming H_1 into the new basis. Already now, there is no simple selection rule or symmetry of the coupling of the states to the leads anymore. We want to use well-established tools of lowest-order perturbation theory for both finding the density matrix of the system and evaluating the current. In order to capture cotunneling by this approach, we perform a Schrieffer-Wolff transformation[13] up to second order, i.e., we take into account all indirect transitions between arbitrary final and initial states of the dot which involve only a *single* intermediate state. This takes the transformed Hamiltonian into the generic form

$$\tilde{H}_I=\sum_{c,d}\alpha_c^\dagger\alpha_d\Bigg[\sum_{Y,Y',\vec{k},\vec{k'}}H^{Y,Y'}_{\vec{k},\vec{k'},c,d}b^{Y\dagger}_{\vec{k}}b^{Y'}_{\vec{k'}}+\sum_{Y,Y',\vec{k},\vec{k'}}H^{Y,Y'}_{\vec{k},\vec{k'},c,d}b^{Y}_{\vec{k}}b^{Y'\dagger}_{\vec{k'}}\Bigg], \tag{6}$$

where the $H^{Y,Y'}_{\vec{k},\vec{k'},c,d}$ are Schrieffer-Wolff amplitudes and $c,d=\pm$ denote the two molecular levels, $\alpha^{(\dagger)}_{c/d}$ the associated molecular operators, and Y,Y' the position of the electrons involved in these processes. Due to the molecular nature of the double-dot eigenstates, all the amplitudes are finite and composed of a huge number of contributions with no particular symmetry. The perturbation-theory formula for this general case can be found, e.g., in Ref. 14 and is worked out in more detail in Refs. 6 and 15. In Eq. (6), we have taken matrix elements in the double-dot eigenbasis only whereas we stick to second-quantized notation in the leads, because this notation readily connects to the formalism used later on.

The stationary density matrix is found using the well-established and controlled Bloch-Redfield theory.[16] This is a systematic technique for deriving generalized master equations within Born approximation in $\tilde{H}_I$, Eq. (6), which includes all relevant non-Markovian parts. This approach has been shown[17] to be numerically equivalent to formally exact path-integral methods for the spin-boson model in the weak-coupling limit. The Redfield equations[18] for the elements of the reduced density matrix ρ in the molecular basis read

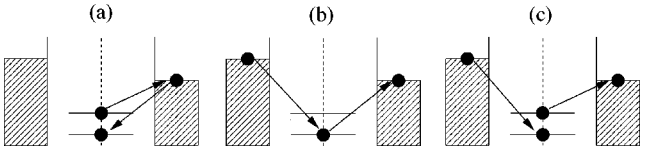

FIG. 2. Examples for relevant processes: (a) a relaxation process without current, (b) current without relaxation (only dephasing), and (c) a process that carries current and also relaxes the system.

$$\dot{\rho}_{nm}(t)=-i\omega_{nm}(t)\rho_{nm}(t)-\sum_{k,l}R_{nmkl}\rho_{kl}(t), \tag{7}$$

where $\omega_{nm}=(E_n-E_m)/\hbar$ are the appropriate energy splittings and R_{nmkl} are the elements of the Redfield tensor. They are composed of golden rule rates involving $\tilde{H}_I$ from Eq. (6). n, m, k, and l can be either $+$ (molecular excited state) or $-$ (molecular ground state). The E's are the eigenenergies of the two molecular states. Due to the lack of symmetry, this leads to a huge number of processes contributing to each term.[15] We are only interested in stationary solutions here. A full treatment of the simple case with $\gamma=0$ can be found in Ref. 6.

The current is derived from the standard formula[19]

$$I(t)=-e\frac{i}{\hbar}\int_{-\infty}^{t}dt'\langle[\dot{N}_L(t),\tilde{H}_I(t')]\rangle, \tag{8}$$

where N_L is the particle number operator on the left dot in the interaction representation and the transformed interaction Hamiltonian $\tilde{H}_I$ from Eq. (6) is also taken in the interaction picture. Carrying out the integration in Eq. (8) and rotating back to the Schrödinger picture, we get a time-independent expression for the current I. Using the stationary occupation probability of the molecular ground ($\rho_{--,\rm st}$) or excited state ($\rho_{++,\rm st}$), we obtain for the expectation value of the stationary current

$$I_{\rm st}={\rm tr}(\rho_{\rm st}I)=\rho_{++,\rm st}I_{++}+\rho_{--,\rm st}I_{--}, \tag{9}$$

where we find from balancing relaxation processes in the Bloch-Redfield equation, Eq. (7),

$$\rho_{++,\rm st}=\frac{R_{++--}}{R_{++--}-R_{++++}},\quad \rho_{--,\rm st}=\frac{R_{--++}}{R_{--++}-R_{----}}. \tag{10}$$

The current amplitudes I_{++} and I_{--} in Eq. (9) are of the same form as the contributions to the Redfield tensor. We emphasize that the choice of processes from all possibilities is very distinct. As an example, Fig. 2 displays a variety of possible processes in such a double-dot system. Processes of the type displayed in Fig. 2(a) contribute to the relaxation but do not carry current, (b) shows a process which carries current but does not relax the state, and (c) relaxes *and* carries

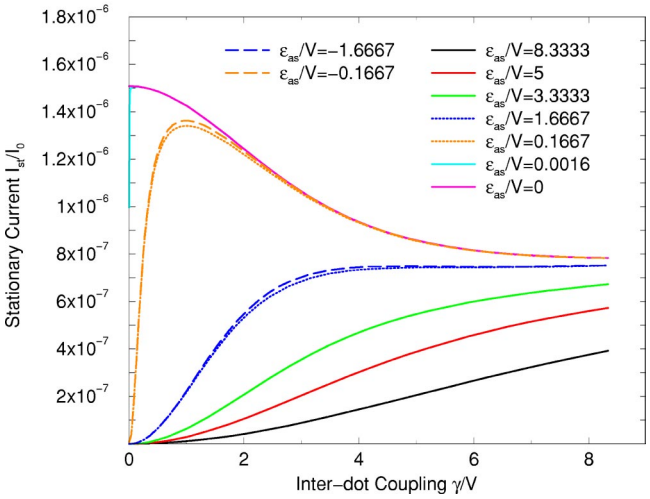

FIG. 3. Stationary current I_{st}/I_0 for different ε_{as}/V as a function of the coupling γ/V (with $T=140$ mK, $V=5.170$ μV and $\mu_{av}=(\mu_R+\mu_L)/2=75.832$ μeV and $\Gamma=1$ GHz).

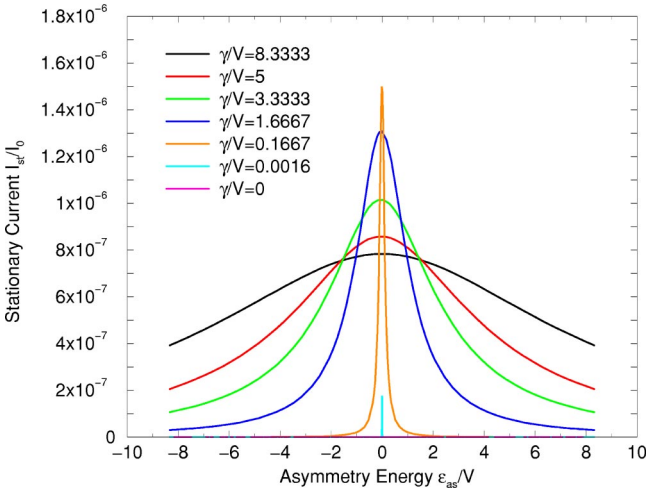

FIG. 4. Stationary current I_{st}/I_0 for different values of γ/V as a function of the asymmetry energy ε_{as}/V (with $T=140$ mK, $V=5.170$ μV and $\mu_{av}=75.832$ μeV and $\Gamma=1$ GHz).

current. The phase information of the quantum state is lost in all three pictures of Fig. 2. Consequently, one must not confuse cotunneling rates with relaxation rates.

We now turn to the discussion of the results. All internal energies ε_{as} and γ are normalized in units of the bias voltage V, the stationary current I_{st} in terms of $I_0=e\Gamma$.

In Fig. 3, the current at fixed bias voltage as a function of the interdot coupling is shown. The sign of ε_{as} plays a role, as one can see above, for an intermediate ε_{as} regime. This effect is more pronounced in $I(V)$, see Ref. 15. Close to $\gamma=0$, the curves all turn to zero because at that point the dots are disconnected and no current can flow. However, a number of curves, the ones with $\varepsilon_{as}/V<1$, exhibit an intermediate maximum at low γ next to a very sharp minimum at $\gamma=0$, which sometimes is hardly resolved. At high $\gamma\gtrsim V$, the stationary current saturates into a value, which for our parameters turns out to be about $I_{0,st}/I_0=7.5\times10^{-7}$. Remarkably, this is half the value of the current at the aforementioned low-γ maximum. This is the central result of this paper.

These regimes can be classified in terms of the level splitting 2δ:[20] At $V<2\delta$, the energy V supplied from the leads is only sufficient to use one of the molecular states for transport (elastic cotunneling) whereas at $V>2\delta$, both states participate and also inelastic processes contribute, i.e., there is a second current channel, which carries the same contribution of I_0. The crossover naturally occurs at $\gamma=\sqrt{V^2/4-\varepsilon_{as}^2}$, which can only be reached if $\varepsilon_{as}/V<1/2$. As long as γ is not too low, the coupling to the leads is the limiting element for the current flow. Only if $\gamma<\varepsilon_{as}$, the double-dot eigenstates become localized and the interdot coupling becomes the current bottleneck. Consequently, associated dips have a half-width of ε_{as} for low temperatures and bias voltages and can thus be extremely narrow. We would like to remark that the notion of transport ''channels'' is appropriate here because cotunneling is a coherent transport process.

Figure 4 shows the dependence of the stationary current on ε_{as}/V. It confirms the interpretation of Fig. 3. The plot is only weakly asymmetric to $\varepsilon_{as}/V=0$. At zero asymmetry, $\varepsilon_{as}/V=0$, the condition for charge transport is ideal, $\sqrt{V^2/4-\varepsilon_{as}^2}$ has its maximum and therefore the current is only governed by the interdot coupling γ/V, resulting in a zero-asymmetry maximum.

Still, all three transport regimes can be recognized in Fig. 4. The $\gamma/V=0$ curve shows that the stationary current I_{st}/I_0 is exactly zero as expected. For growing, but small values of γ/V, the maximum at $\varepsilon_{as}/V=0$ reaches the highest value $I_{st}/I_0=2I_{0,st}/I_0$ at about 1.5×10^{-6} (like in Fig. 3), corresponding to two open transport channels (elastic *and* inelastic). If we raise γ/V further, the height of the peak goes down again and saturates at $I_{st}/I_0=I_{0,st}/I_0\approx7.5\times10^{-7}$, corresponding to only the elastic channel being open.

The three transport regimes are summarized in Fig. 5: (i)

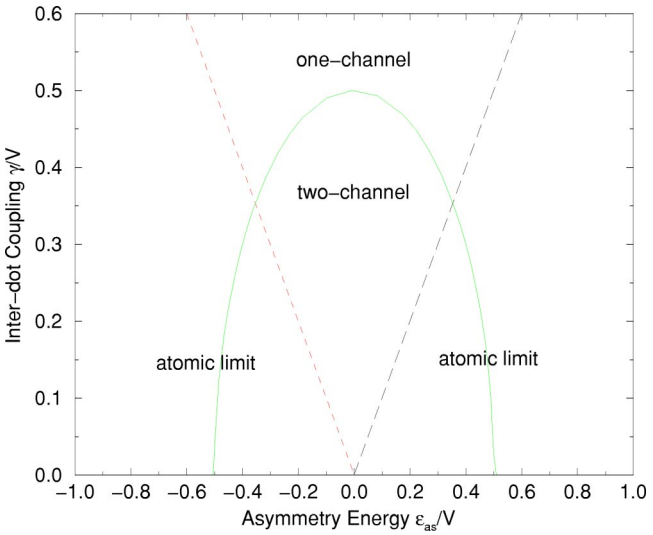

FIG. 5. Limits for the three transport regimes with $V=5.170$ μV.

the atomic limit (no transport) $\gamma < \varepsilon_{as}$, (ii) the two-channel case (inelastic cotunneling) $\varepsilon_{as} < \gamma < \sqrt{V^2/4 - \varepsilon_{as}^2}$, and (iii) the one-channel case (elastic cotunneling) $\gamma > \sqrt{V^2/4 - \varepsilon_{as}^2}$.

These conditions show that indeed cotunneling can be used as a tool for investigating the energy spectrum of an undisturbed artificial molecule.[4] The crossover between the elastic and the inelastic cotunneling in dependence of the applied bias voltage has recently been observed.[12] A similar conclusion was found in Ref. 20.

Although the notion of (elastic and inelastic) cotunneling was already introduced very early,[11] its consequences for realistic quantum dot systems have only been discussed very recently,[20] along with detailed and accurate experiments on small semiconductor quantum dots[4,12] becoming available. The sharp crossover between elastic and inelastic cotunneling, which we discuss, has been identified in a vertical quantum dot[12] by changing the transport voltage. Reference 4 studies cotunneling in a parallel double-dot topology, using again cotunneling and the elastic-to-inelastic crossover as a spectroscopic tool and tuning the interdot coupling *in situ*. In both cases, the narrow regime of decoupled dots would not have been accessible through a conductance measurement. Some of the experimental issues have been theoretically addressed in Ref. 20. In that case, however, the behavior of a single multilevel dot system was modeled with phenomenological couplings to the leads, whereas we take a realistic model and only by this manage to predict effects which, e.g., depend on the serial dot topology of the sample. Note that parts of the double-dot literature focus on phonon/photon assisted transport (see, e.g., Refs. 21 and 22 for experiments and Refs. 23 and 24 for theory). Unlike Ref. 25, we concentrate on the Coulomb blockade regime and do not consider cotunneling at resonance. In Ref. 26, a different approach to the problem was developed, in which the master equation is carried to second order instead of using a Schrieffer-Wolff transformation, and a few setups simpler than ours are studied. Our approach does not require the molecule to be artificial, in principle, it can be applied to ''real'' molecules.[27] In contrast to the approach in Ref. 28, it permits to take into account charging effects, however, the Schrieffer-Wolff transformation is clearly a laborious step for larger systems.

To conclude, we analyzed the stationary coherent cotunneling current I_{st} through a double quantum dot system or artificial molecule. As a function of the interdot coupling strength it displays a rich, nonmonotonic structure, which enables us to perform ''molecular cotunneling spectroscopy.'' Strikingly, we have shown that at fixed bias voltage, the current is highest, if the dots are weakly to intermediately connected, such that the interdot coupling is at least as strong as the coupling to the leads, but the splitting of the molecular wave functions is still smaller than the transport voltage.

We thank J. von Delft, J. König, L. Borda, M. Sindel, A.W. Holleitner, A.K. Hüttel, and E.M. Höhberger for clarifying discussions. We acknowledge financial support from ARO, Contract No. P-43385-PH-QC.

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[1] R.C. Ashoori, Nature (London) **379**, 413 (1996).
[2] F.R. Waugh *et al.*, Phys. Rev. Lett. **75**, 705 (1995).
[3] R.H. Blick *et al.*, Phys. Rev. Lett. **80**, 4032 (1998).
[4] A.W. Holleitner *et al.*, Science **297**, 70 (2002).
[5] R.H. Blick and H. Lorenz, Proc. ISCAS **2000**, II245 (2000).
[6] U. Hartmann and F.K. Wilhelm, Phys. Status Solidi B **233**, 385 (2002).
[7] L.P. Kouwenhoven *et al.*, in *Mesoscopic Electron Transport*, Vol. 345 of *NATO Advanced Studies Institute, Series E: Applied Sciences*, edited by L.L. Sohn, L.P. Kouwenhoven, and Gerd Schön (Kluwer Academic, Dordrecht, 1997), p. 105.
[8] W.G. van der Wiel *et al.*, Rev. Mod. Phys. **75**, 1 (2003).
[9] T.H. Stoof and Yu.V. Nazarov, Phys. Rev. B **53**, 1050 (1996).
[10] See e.g., L. Borda *et al.*, Phys. Rev. Lett. **90**, 026602 (2003).
[11] D. V. Averin and Yu. V. Nazarov, in *Single Charge Tunneling*, Vol. 294 of *NATO Advanced Studies Institute, Series B: Physics*, edited by H. Grabert and M. H. Devoret (Plenum, New York, 1992), pp. 217–247.
[12] S. De Franceschi *et al.*, Phys. Rev. Lett. **86**, 878 (2001).
[13] J.R. Schrieffer and P.A. Wolff, Phys. Rev. **149**, 491 (1966).
[14] C. Cohen-Tannoudij, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (Wiley, New York, 1992).
[15] U. Hartmann, Master's thesis, University of Bonn, 2002; U. Hartmann and F. K. Wilhelm (unpublished).
[16] P.N. Agyres and P.L. Kelley, Phys. Rev. **A134**, 98 (1964).
[17] L. Hartmann, I. Goychuk, M. Grifoni, and P. Hänggi, Phys. Rev. E **61**, R4687 (2000).
[18] U. Weiss, *Quantum Dissipative Systems*, 2nd ed. (World Scientific, Singapore, 1999).
[19] G. D. Mahan, *Many-Particle Physics*, 3rd ed. (Kluwer Academic, New York, 2000).
[20] M.R. Wegewijs and Yu.V. Nazarov, cond-mat/0103579 (unpublished).
[21] H. Qin, A.W. Holleitner, K. Eberl, and R.H. Blick, Phys. Rev. B **64**, 241302 (2001).
[22] T.H. Oosterkamp *et al.*, Nature (London) **395**, 873 (1998).
[23] B.L. Hazelzet, M.R. Wegewijs, T.H. Stoof, and Yu.V. Nazarov, Phys. Rev. B **63**, 165313 (2001).
[24] T. Brandes and B. Kramer, Phys. Rev. Lett. **83**, 3021 (1999).
[25] J. König, H. Schoeller, and G. Schön, Phys. Rev. Lett. **78**, 4482 (1997).
[26] E.V. Sukhorukov, G. Burkard, and D. Loss, Phys. Rev. B **63**, 125315 (2001).
[27] H.B. Weber *et al.*, Chem. Phys. (to be published).
[28] J. Heurich, J.C. Cuevas, W. Wenzel, and G. Schön, Phys. Rev. Lett. **88**, 256803 (2002).