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Breaking of phase symmetry in nonequilibrium Aharonov-Bohm oscillations through a quantum dot

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Linear-response conductance of a two-terminal Aharonov-Bohm (AB) interferometer is an even function of magnetic field. This *phase symmetry* is not expected to hold beyond the linear-response regime and in simple AB rings the phase of the oscillations changes smoothly (almost linearly) with voltage bias. However, in an interferometer with a quantum dot in its arm, tuned to the Coulomb blockade regime, experiments indicate that phase symmetry seems to persist even in the nonlinear regime. In this paper we discuss the processes that break AB phase symmetry and show that breaking of phase symmetry in such an interferometer is possible only after the onset of inelastic cotunneling, i.e., when the voltage bias is larger than the excitation energy in the dot. The asymmetric component of AB oscillations is significant only when the contributions of different levels to the symmetric component nearly cancel out (e.g., due to different parity of these levels), which explains the sharp changes in the AB phase. We show that our theoretical results are consistent with experimental findings.

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I. INTRODUCTION

The Aharonov-Bohm (AB) effect allows for studying the transmission phase through a mesoscopic structure, e.g., a quantum dot (QD), by placing it in one of the arms of an AB interferometer.[1,2] In a two-terminal interferometer the phase of the AB oscillations in the linear-response conductance can only assume the values 0 or π (i.e., the oscillations have either maximum or minimum at zero magnetic field), even though the transmission phase through the QD can change continuously. This *phase symmetry*, i.e., the property that the linear-response conductance of a two-terminal device is an even function of magnetic flux, can be understood within a one-particle picture[3] and is, in fact, a manifestation of more general linear-response Onsager-Büttiker symmetries.[4,5] Deviations from phase symmetry in two-terminal devices in the nonlinear regime have been studied theoretically,[6–8] as well as in experiments on AB cavities[9] and AB rings.[10] The resulting phase of the AB oscillations changes smoothly (almost linearly) with increasing voltage bias.[10]

Rather puzzlingly, a recent experiment,[11] which studied a voltage-biased AB interferometer with Coulomb blockaded QDs in its arms, observed AB oscillations which remained practically symmetric. The phase of the oscillations changed with voltage bias, V_{sd}, in a highly nonmonotonous fashion: it remained close to 0 or π but switched abruptly between these two values as a function of the bias voltage, with the first switching occurring when the voltage about equal to the level spacing to the first excited state Δ, i.e., near the onset of inelastic cotunneling.

Indeed, breaking of the phase symmetry in the regime of inelastic cotunneling have not been addressed theoretically thus far. In particular, the finite bias threshold for the inelastic cotunneling renders inapplicable the methods based on expansion in powers of the voltage bias V_{sd},[7] and thus cannot explain the experimental observations. In this paper we address the phase asymmetry of AB oscillations in a QD interferometer with a Coulomb blockaded dot by systematically analyzing transport processes of different order in lead-to-lead tunnel coupling. We demonstrate that the bias dependence of the AB phase is highly nonmonotonous. In particular, (i) the oscillations indeed remain symmetric up to the onset of inelastic cotunneling ($eV_{sd} \simeq \Delta$) (i.e., with AB phase 0 or π), in agreement with experiments; (ii) with onset of inelastic cotunneling, AB oscillations acquire nonzero asymmetric component, which however is usually smaller than the symmetric component, the oscillations thus remaining nearly symmetric; (iii) the asymmetric component may become dominant if the contributions of different levels to even AB oscillations nearly cancel out (e.g., due to different parity of these levels).[12] The theoretical findings are supported by the in-depth analysis of the experimental data of Ref. 11.

II. THEORETICAL FORMULATION

We consider an AB interferometer schematically shown in Fig. 1(a). One arm of the interferometer contains a QD which is assumed to be in Coulomb blockade regime. The current

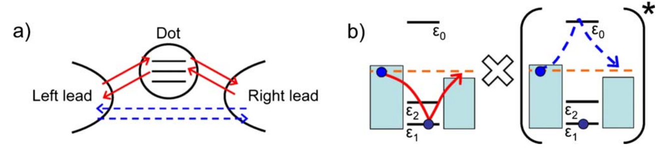

FIG. 1. (Color online) (a) Schematic representation of the device studied in this paper. Solid red and dash blue arrows show cotunneling processes and direct lead-to-lead tunneling, respectively. (b) Example of a lowest-order cotunneling process contributing to AB oscillations: the dot electron tunnels to the right lead and an electron from the left tunnels to the dot, interfering with the process where an electron moves from left to right through the open arm.

can flow either by means of cotunneling via the QD or by direct lead-to-lead tunneling through the open arm of the interferometer,[13] whereas the number of electrons occupying the QD does not change.

We describe the system by Hamiltonian $H=H_L+H_R+H_D+V+W$, where $H_\mu=\Sigma_E E c^+_{\mu E}c_{\mu E}$ is the Hamiltonian of electrons in lead $\mu=L,R$; E labels energy states within one lead. $H_D=\Sigma_\beta \epsilon_\beta d^+_\beta d_\beta$ is the Hamiltonian of the QD, which contains only one electron and has energy levels ϵ_β. $c_{\mu E}$ destroys a lead electron in state μE, d_β destroys QD state β.[14]

W and V describe, respectively, electron transitions between the leads through the open arm or through the arm that contains the QD. Due to the Coulomb blockade, the number of electrons in the QD after the electron transfer remains unchanged but the process can be accompanied by virtual change in the QD state. These terms in the Hamiltonian are given by

$$W=\sum_{\mu E}\sum_{\mu' E'} W_{\mu;\mu'}e^{i\phi_{\mu\mu'}}c^+_{\mu E}c_{\mu' E'} \tag{1a}$$

$$V=\sum_{\beta,\beta'}\sum_{\mu E}\sum_{\mu' E'} V^{\beta;\beta'}_{\mu;\mu'}d^+_\beta c^+_{\mu E}c_{\mu' E'}d_{\beta'}, \tag{1b}$$

where $W_{\mu;\mu'}$ and $V^{\beta;\beta'}_{\mu;\mu'}$ are real and ϕ is the magnetic flux through the interferometer ($\phi_{RL}=-\phi_{LR}=\phi$, $\phi_{LL}=\phi_{RR}=0$).[14]

We calculate the lead-to-lead current perturbatively in powers of V and W. This calculation, described in detail in Sec. 2 of Appendix, is similar to that used, e.g., by Appelbaum for Kondo problem[15] (identical results were obtained using the Keldysh formalism). The method consists of calculating the quantum-mechanical probability for an electron to be transferred from one lead to another, which is then averaged over initial states with the correct weights and summed over all final and intermediate virtual states. The essential modification of this approach necessary for a nonequilibrium problem is the correct choice of zeroth-order level occupation numbers.[16] These occupation numbers, $P_\beta(V_{sd})$, although of zero order in the tunneling elements V,W, are dependent on V_{sd}. In particular, at low bias, only the population of the ground state of the QD, $P_1(V_{sd})$, significantly differs from zero. When the bias exceeds the threshold for onset of inelastic cotunneling, $V_{sd}>\Delta\equiv\epsilon_2-\epsilon_1$, the populations of excited QD states start to grow. The dependence of these populations on the source-drain bias was studied in Ref. 12.

III. BREAKING OF PHASE SYMMETRY

It is easy to see that the second-order processes contributing to the AB oscillations (which necessarily involve one tunneling amplitude through the open arm, W, and one through the dot, V), such as the one depicted in Fig. 1(b) (where ϵ_0 represents the open arm), are necessarily symmetric with respect to magnetic field. The asymmetric AB oscillations appear when we account for higher-order tunneling processes. Typical third-order contributions to AB oscillations are depicted in Fig. 2. As an example, the probabilities of the processes shown in Figs. 2(a) and 2(b), are, respectively,

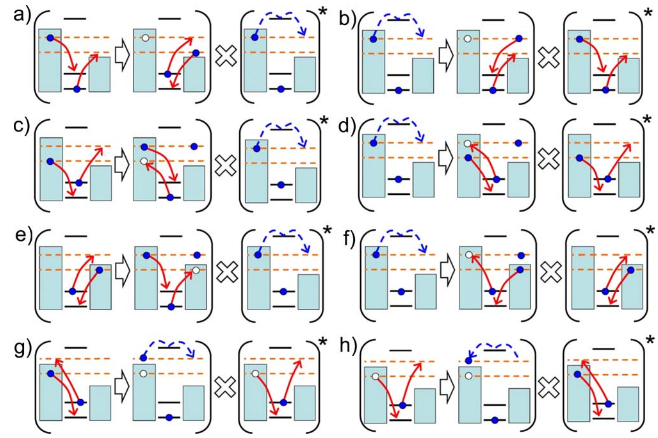

FIG. 2. (Color online) Examples of different third-order processes, whose real parts contribute to the current: (a) and (b) [or (c) and (d)] are two processes whose contributions to odd AB oscillations mutually cancel out; processes (a) and (c) are elastic, whereas (b) and (d) are inelastic. (e) [or (g)] is an example of an *elastic* (*inelastic*) third-order process which gives nonzero contribution to the odd AB oscillations. The other process constructed from the same matrix elements and beginning from the same initial state, (f) [or (h)], does not contribute to the current.

$$4\pi\Re\left[\frac{(W_{R;L}e^{i\phi})^* V^{1;2}_{R;R}V^{2;1}_{R;L}}{\epsilon_1+E_L-\epsilon_2-\tilde{E}_R+i0^+}\right]\delta(E_L-E_R), \tag{2a}$$

$$4\pi\Re\left[\frac{(V^{2;1}_{R;L})^* V^{2;1}_{R;R}W_{R;L}e^{i\phi}}{E_L-E_R+i0^+}\right]\delta(E_L+\epsilon_1-\tilde{E}_R-\epsilon_2), \tag{2b}$$

($\Re$ represents the real part). These factors consist of the second-order tunneling amplitude (which contains the energy denominator) multiplied by the complex conjugate of the first-order tunneling amplitude: this is reflected in the obvious fashion in Fig. 2, upon which the following discussion is built. There are also processes (not shown here) in which instead of an electron one considers tunneling of a hole.

In order to obtain their contributions to the current, the probabilities in Eq. (2) are multiplied by the factor $P_1(V_{sd})f_L(E_L)[1-f_R(E_R)][1-f_R(\tilde{E}_R)]$ (which also limits possible intermediate states) and integrated over E_L,E_R and $\tilde{E}_R$.

The asymmetric contribution to AB oscillations results only from the imaginary part of the denominators in Eqs. (2), which we treat according to prescription $1/(E+i0^+)=1/E-i\pi\delta(E)$.[17] The delta function means that only processes in which the *intermediate state lies on the same energy shell with the initial and the final states*, which for our example means that $E_L+\epsilon_1=E_R+\epsilon_1=\tilde{E}_R+\epsilon_2$, contribute to AB oscillations odd in magnetic field.

The asymmetric contribution due to the process [Eq. (2a)] is thus given by

$$-2W_{R;L}V^{1;2}_{R;R}V^{2;1}_{R;L}\delta(\epsilon_1+E_L-\epsilon_2-\tilde{E}_R)\delta(E_L-E_R)\sin\phi. \tag{3}$$

On the other hand, the asymmetric contribution of the process [Eq. (2b)] is given by the exact same expression but with the opposite sign and thus the asymmetry contribution is canceled between these two processes. This is no surprise. The first process [Fig. 2(a)] corresponds to the dot starting with an electron in the ground state. Then this electron tunnels to the right and an electron from the left tunnels to the excited state, then the electron tunnels from the excited state to the right lead and another electron tunnels from the same lead to the ground state ending at the same initial state but one electron transferred from left to right. This probability amplitude interferes with the amplitude of one-electron tunneling directly through the other arm from left to right. The second process [Fig. 2(b)] starts with the same initial state, and involves an electron tunneling through the other arm to the right lead, and then an electron from the right lead tunneling to the excited state, while the ground-state electron tunnels to the right lead. This amplitude, which again involves one electron moving from left to right, interferes with the amplitude where the dot electron tunnels to the right and an electron from the left tunnels to the excited state. These two processes, which have the same weight as they start from the same initial configuration, involve the exact same matrix elements, but effectively correspond to electron traversing the AB ring in opposite directions, thus leading to the cancellation of the term odd in magnetic field. Similar cancellation occurs for the processes starting with the QD in its exited state, Figs. 2(c) and 2(d).

However, let us examine the process shown in Fig. 2(e). The process that should cancel its asymmetric contribution is depicted in Fig. 2(f). This latter process, however, does not contribute to the current, as it describes electron backscattered into the same lead. Thus, the contribution of the elastic process in Fig. 2(e) gives rise to AB oscillations odd in magnetic field. Figures 2(g) and 2(h) provide an example of a similar noncanceling inelastic process.

The distinctive feature of the processes in Figs. 2(e) and 2(g) is that prior to electron transfer from left to right, an electron is being excited to a state within the same lead. When this part of the process is singled out as a one-particle amplitude in the other process made up of the same elements and beginning from the same initial state, Figs. 2(f) and 2(h), we obtain processes which only involve excitation within the same lead, and thus do not contribute to the current, i.e., do not contribute to the measured AB oscillations.

Since such a preliminary excitation is possible only when the QD is initially in its excited state, whose population differs from zero only when $eV_{sd}>\Delta$, *breaking of the phase symmetry may happen only after the onset of inelastic cotunneling*.

The asymmetric contribution to AB oscillations is of higher order in the lead-to-lead coupling than the symmetric contribution. Thus, the asymmetry should be weak everywhere, except the bias values where second-order processes vanish due to canceling contributions from different levels, i.e., when phase switching occurs.[12] Overall, this means that *the phase of AB oscillations is not a monotonous function of bias: it is usually very close to* $0,\pi$ *but deviates significantly from these values when phase switching occurs*.

IV. DISCUSSION AND COMPARISON TO THE EXPERIMENT

Here we report calculations with a three level dot, similar to that used in Ref. 12 in connection to the experiments of Ref. 11: the levels have alternating parity and different strength of coupling to the leads.

The AB component of differential conductance obtained within the perturbation framework described above is shown in the upper left panel of Fig. 3. One can see that the phase of the AB oscillations changes between 0 and π. The lower left panel of Fig. 3 depicts the asymmetric component of AB oscillations extracted from the data shown in the upper left. The right part of Fig. 3 presents, respectively, total (upper panel) and asymmetric (lower panel) contributions to AB oscillations as obtained from the experimental data of Ref. 11.

In both theoretical and experimental color plots one can observe several important features: (i) the phase of AB oscillations switches sharply between values close to 0 and π;[11,12] (ii) in the figures showing total AB signal any significant asymmetry is seen only in the regions corresponding to phase switching, e.g., close to $V_{sd}=\pm 0.2$ mV in the upper part of Fig. 3; (iii) the asymmetric component of AB oscillations is zero for bias below the onset of inelastic cotunneling but nonzero essentially everywhere above this onset.

In order to illustrate the last point we show in Fig. 4 the mean differential conductance through the interferometer together with the power of the asymmetric component, calculated as $P(V_{sd})=\sqrt{\int_{B_{min}}^{B_{max}}dBG_{asym}^2(B,V_{sd})/(B_{max}-B_{min})}$, where $G_{asym}(B,V_{SD})$ is the asymmetric component of the differential conductance. For the theoretical model limits B_{min} and B_{max} are restricted to one period of AB oscillations. At the onset of inelastic cotunneling the differential conductance exhibits a jump, which is due to increase in the available conductance processes. We see that the power of the asymmetric component mimics the onset of inelastic cotunneling, which confirms our theoretical predictions. The nonzero value of the asymmetric AB oscillations before the onset of inelastic cotunneling in experimental data most likely results from finite extension of the electron density throughout the device (i.e., not all localized to QD). In this case the electric potential within the device becomes a function of magnetic field, which leads to asymmetry of AB oscillations,[7] which however grows smoothly with the bias voltage.[10]

A noticeable difference between the experimental and theoretical data is that the asymmetric component of the experimental signal seems mainly even in bias while the signal is mostly odd in the calculations. Our theoretical study showed that the even bias component is nonzero only when the dot levels are not symmetrically coupled to the leads. A proper treatment of this even contribution requires taking higher-order terms in the expansion of the current in lead-to-lead tunneling matrix elements, which is beyond our current calculation. Therefore we limited our theoretical calculation to the symmetric voltage component and we chose the parameters that make the theoretical curves resemble the experimental ones for positive bias side. Another difference between the theory and the experiments is that in the

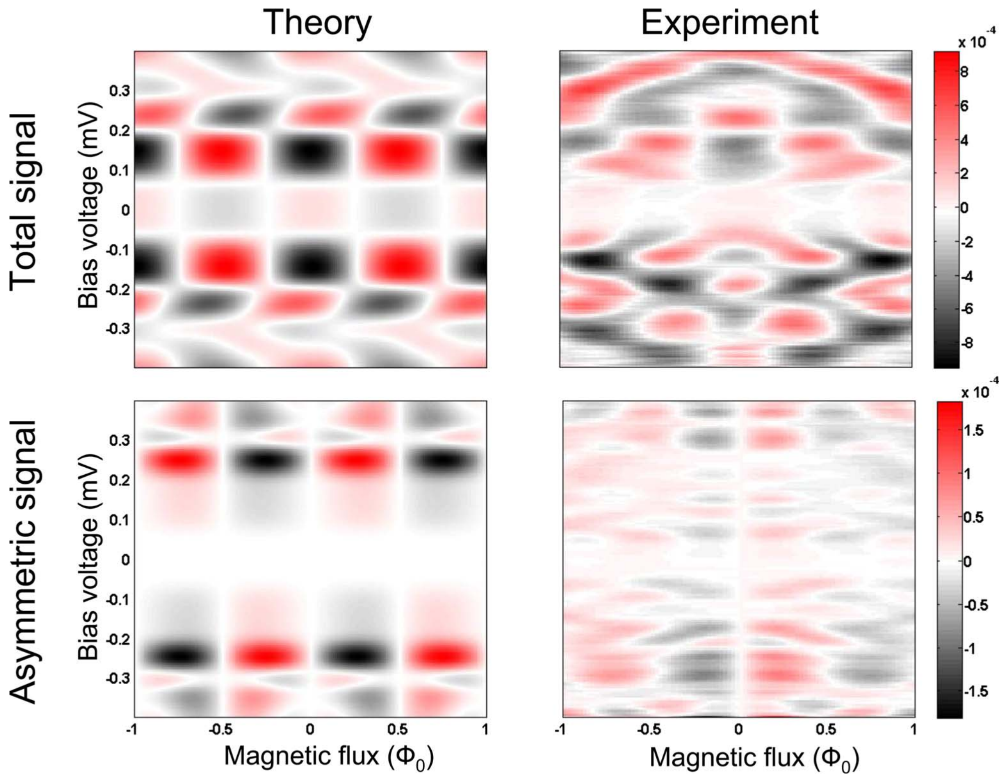

FIG. 3. (Color online) Color plots of the differential conductance obtained from the theoretical model presented here (left panels) and from the experimental data of Ref. 11 (right panels). The upper and lower panels show, respectively, full and asymmetric components of the conductance.

experimental data the asymmetric component of the AB conductance changes sign many times. This feature may be the result of the weakness of the asymmetric AB signal (only about factor of 5 above the noise), the larger number of dot levels in the experiment or the interplay between the coupling strengths of different levels to the leads.[12] Another option is additional influence of the electrostatic AB effect, owing to the finite extension of the interferometer arms.[10]

V. CONCLUSION

We addressed breaking of phase symmetry in a quantum-dot AB interferometer in cotunneling regime. We showed that AB oscillations remain strictly symmetric up to the onset of inelastic cotunneling and discussed the processes responsible for breaking of the phase symmetry above this onset. As asymmetric component of AB oscillations is of higher

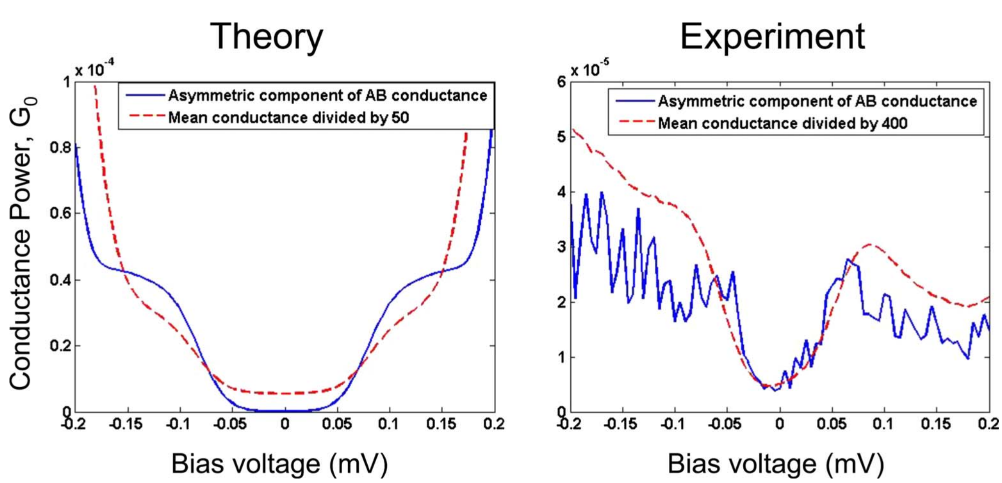

FIG. 4. (Color online) Power of asymmetric AB oscillations and differential conductance (rescaled) for theoretical model (left) and for experimental data (right).

order in lead-to-lead tunneling than the symmetric one, the AB phase remains close to values 0 and π. The exception are the bias values where phase switching occurs and the asymmetric component of AB oscillations becomes dominant. Altogether this results in AB phase changing sharply but continuously between values 0 and π. We show that our theoretical findings are in excellent agreement with the experimental data of Ref. 11.

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APPENDIX: DETAILS OF CALCULATION AND GENERALIZATION TO THE CASE OF MULTICHANNEL LEADS AND ARBITRARY MAGNETIC FIELD DEPENDENCE

1. Theoretical formulation for the case of multichannel leads and arbitrary flux dependence of the matrix elements

We consider an AB interferometer schematically shown in Fig. 1(a). One arm of the interferometer contains a QD which is assumed to be in the Coulomb blockade regime. The current can flow either by means of cotunneling via the QD or by direct lead-to-lead tunneling through the open arm of the interferometer while the number of electrons occupying the QD does not change.

We describe the system by the Hamiltonian $H=H_L+H_R+H_D+\tilde{V}$, where $H_\mu=\Sigma_{\alpha,k}\epsilon_{\mu\alpha k}c^+_{\mu\alpha k}c_{\mu\alpha k}$ is the Hamiltonian of electrons in lead $\mu=L,R$, α labels different lead channels and k labels energy states within one channel. $H_D=\Sigma_\beta\epsilon_\beta d^+_\beta d_\beta$ is the Hamiltonian of the QD, which contains only one electron and has energy levels ϵ_β, $c_{\mu\alpha k}$ destroys a lead electron in state $\mu\alpha k$, and d_β destroys the QD state β.

Electron transitions between the leads are described by the term

$$\tilde{V}(B)=\sum_{\beta,\beta'}\sum_{\mu\alpha k}\sum_{\mu'\alpha'k'}\tilde{V}^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}(B)d^+_\beta c^+_{\mu\alpha k}c_{\mu'\alpha'k'}d_{\beta'}. \tag{A1}$$

In case of the two-arm interferometer studied in this paper one can separate the terms responsible for the transport via each arm

$$\tilde{V}(B)=V(B)+W(B), \tag{A2}$$

where $V(B)$ describes electron tunneling via the QD and $W(B)$ describes tunneling via the open arm of the interferometer. Due to Coulomb blockade, the number of electrons in the QD after the electron transfer remains unchanged, $\Sigma_\beta d^+_\beta d_\beta=1$. If, in addition, we choose to account for the magnetic field only via the AB phase, the decomposition can be written explicitly as

$$\tilde{V}^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}(B)=V^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}+W_{\mu\alpha;\mu'\alpha'}\delta_{\beta;\beta'}e^{i\phi_{\mu\mu'}}, \tag{A3a}$$

$$W(B)=\sum_{\mu\alpha k}\sum_{\mu'\alpha'k'}W_{\mu\alpha;\mu'\alpha'}e^{i\phi_{\mu\mu'}}c^+_{\mu\alpha k}c_{\mu'\alpha'k'} \tag{A3b}$$

$$V(B)=\sum_{\beta,\beta'}\sum_{\mu\alpha k}\sum_{\mu'\alpha'k'}V^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}d^+_\beta c^+_{\mu\alpha k}c_{\mu'\alpha'k'}d_{\beta'}, \tag{A3c}$$

where $W_{\mu\alpha;\mu'\alpha'}$ and $V^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}$ are real, and ϕ is the magnetic flux through the interferometer associated with the magnetic field B ($\phi_{RL}=-\phi_{LR}=\phi$ and $\phi_{LL}=\phi_{RR}=0$).

The decomposition of Eq. (A3) is optional, as the basic statements regarding the phase symmetry breaking in cotunneling transport, obtained in this paper, can be proved using only the properties of the matrix elements $\tilde{V}^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}(B)$ with respect to time reversal. (This is important for interferometers of more complicated geometry, e.g., with more than two arms.) For the choice of the basis states that are invariant under time-reversal transformation (i.e., real), the matrix elements satisfy

$$\tilde{V}^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}(B)=\tilde{V}^{\beta';\beta}_{\mu'\alpha';\mu\alpha}(-B)=[\tilde{V}^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}(-B)]^*. \tag{A4}$$

2. General expression for the current

In order to calculate the lead-to-lead current we employ a perturbative expansion in the powers of $\tilde{V}$, similar to the one used, e.g., by Appelbaum for the Kondo problem.[15] Up to order $\tilde{V}^3$, the current is expressed as $I_\mu(B)=I_{\mu\leftarrow\bar{\mu}}(B)-I_{\bar{\mu}\leftarrow\mu}(B)$, where

$$I_{\mu\leftarrow\bar{\mu}}(B)=\frac{e}{2\pi\hbar}\int d\epsilon\int d\epsilon'\left[\sum_{\beta\alpha;\beta'\alpha'}T^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(\epsilon,\epsilon',B)\right]\delta(\epsilon_\beta+\epsilon-\epsilon_{\beta'}-\epsilon')P_{\beta'}f_{\bar{\mu}}(\epsilon')[1-f_\mu(\epsilon)], \tag{A5a}$$

$$T^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(\epsilon,\epsilon',B)\approx(2\pi)^2N_\mu N_{\bar{\mu}}\{|\tilde{V}^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(B)|^2+2\Re[(\tilde{V}^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(B))^*A^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(\epsilon,\epsilon',B)]\}, \tag{A5b}$$

$$A^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(\epsilon,\epsilon',B)=\sum_{\mu''}\sum_{\beta''\alpha''}N_{\mu''}\int d\epsilon''\left\{\frac{\tilde{V}^{\beta;\beta''}_{\mu\alpha;\mu''\alpha''}(B)\tilde{V}^{\beta'';\beta'}_{\mu''\alpha'';\bar{\mu}\alpha'}(B)}{\epsilon_{\beta'}+\epsilon'-\epsilon_{\beta''}-\epsilon''+i\eta}[1-f_{\mu''}(\epsilon'')]-\frac{\tilde{V}^{\beta;\beta''}_{\mu''\alpha'';\bar{\mu}\alpha'}(B)\tilde{V}^{\beta'';\beta'}_{\mu\alpha;\mu''\alpha''}(B)}{\epsilon_{\beta'}+\epsilon''-\epsilon_{\beta''}-\epsilon+i\eta}f_{\mu''}(\epsilon'')\right\}. \quad \text{(A5c)}$$

In these equations N_μ is the density of states in lead μ, whereas $f_\mu(\epsilon)=1/\{\exp[(\epsilon-\zeta_\mu)/(k_BT)]+1\}$ is the Fermi distribution function in this lead. The difference of the lead chemical potentials, ζ_μ, is given by the source-drain bias, $eV_{sd}=\zeta_L-\zeta_R$.

The essential modification to the perturbative approach,[15] necessary in a nonequilibrium problem, is the correct choice of zero-order level occupation numbers, which can be done on the basis of the second-order transition rates.[16] These occupation numbers, $P_\beta(V_{sd})$, although of zero order in $\tilde{V}(B)$, are dependent on the source-drain bias, V_{sd}. In particular, at low bias V_{sd}, only the population of the ground state of the QD, $P_1(V_{sd})$, significantly differs from zero. The situation changes when the bias exceeds the threshold for the *onset of inelastic cotunneling*, $V_{sd}>\Delta=\epsilon_2-\epsilon_1$, after which the populations of excited QD states start to grow. The dependence of these populations on the source-drain bias was studied in more detail in Ref. 12.

3. Identifying the processes responsible for breaking of the phase symmetry

As readily follows from Eq. (A4), the current at order $\tilde{V}^2$ is symmetric in magnetic field since

$$|\tilde{V}^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(B)|^2=|\tilde{V}^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(-B)|^2. \quad \text{(A6)}$$

Any deviations from the phase symmetry come about due the second term in Eq. (A5b). We expand the denominators in the second-order tunneling amplitude, Eq. (A5c), according to the standard prescription, $1/(\epsilon+i\eta)=1/\epsilon-i\pi\delta(\epsilon)$, and take into account that, due to Eq. (A4),

$$[\tilde{V}^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(B)]^*\tilde{V}^{\beta;\beta''}_{\mu\alpha;\mu''\alpha''}(B)\tilde{V}^{\beta'';\beta'}_{\mu''\alpha'';\bar{\mu}\alpha'}(B)=\tilde{V}^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'}(-B)\times[\tilde{V}^{\beta;\beta''}_{\mu\alpha;\mu''\alpha''}(-B)\tilde{V}^{\beta'';\beta'}_{\mu''\alpha'';\bar{\mu}\alpha'}(-B)]^*, \quad \text{(A7)}$$

i.e., the real part of this expression is even in magnetic field. (Here and below we discuss only the first, "electron," term in Eq. (A5c) but the second, "hole," term can be treated similarly.) Thus, the contribution to AB oscillations odd in magnetic field may result only from the terms proportional to the delta functions, i.e., it comes from the processes in which the intermediate state lies on the same energy shell with the initial and the final states, $\epsilon+\epsilon_\beta=\epsilon'+\epsilon_{\beta'}=\epsilon''+\epsilon_{\beta''}$.[17]

We now need to distinguish three kinds of processes: (i) elastic process in which no change in the QD state occurs, i.e., $\beta=\beta'=\beta''$; (ii) elastic processes in which the intermediate state of the QD is different from its initial and final states, i.e., $\beta=\beta'\neq\beta''$; and (iii) inelastic processes, $\beta\neq\beta'$.

For type (i) processes we take advantage of the two lead geometry (i.e., that μ'' is limited to $\mu,\bar{\mu}$) to prove that, e.g., for $\mu''=\mu$

$$\sum_{\alpha\alpha'\alpha''}[\tilde{V}^{\beta;\beta}_{\mu\alpha;\bar{\mu}\alpha'}(B)]^*\tilde{V}^{\beta;\beta}_{\mu\alpha;\mu\alpha''}(B)\tilde{V}^{\beta;\beta}_{\mu\alpha'';\bar{\mu}\alpha'}(B)=\sum_{\alpha\alpha'\alpha''}[\tilde{V}^{\beta;\beta}_{\mu\alpha;\bar{\mu}\alpha'}(-B)]^*\tilde{V}^{\beta;\beta}_{\mu\alpha;\mu\alpha''}(-B)\tilde{V}^{\beta;\beta}_{\mu\alpha'';\bar{\mu}\alpha'}(-B). \quad \text{(A8)}$$

A similar relation holds for $\mu''=\bar{\mu}$. Therefore these processes do not contribute to odd AB oscillations.

The same argument cannot be applied to the summation over β,β',β'' due to the presence of factor P_β and Fermi functions in this summation. (These factors, however, can be taken out of the summation in case of degenerate levels, which are equally populated.)

Before the onset of inelastic cotunneling the inelastic processes, (iii), are prohibited by energy conservation (however, let us point out that these processes will also contribute to AB oscillations since they are indistinguishable from lower-order inelastic processes). The processes of type (ii) are possible but the Fermi factors in Eq. (A5c) mean that the intermediate states on the same energy shell with the initial and the final states are not available. Thus, *no breaking of phase symmetry is possible before the inelastic cotunneling sets on.*

4. Asymmetric term for a two-arm interferometer

We now quote the result for the case considered in the main text of this paper, i.e., when the decomposition of Eq. (A3) applies. The lowest-order contribution to AB oscillations is proportional to the oscillating part of $|\tilde{V}^{\beta;\beta'}_{\mu\alpha;\mu'\alpha'}(\phi)|^2$ which is

$$2V^{\beta;\beta}_{\mu\alpha;\bar{\mu}\alpha'}\delta_{\beta,\beta'}W_{\mu\alpha;\bar{\mu}\alpha'}\cos\phi. \quad \text{(A9)}$$

The Kroneker symbol $\delta_{\beta,\beta'}$ reflects the fact that the processes changing the state of the QD do not contribute to the leading term in AB oscillations.[18] The cosine function in Eq. (A9), $\cos\phi$, tells us that these oscillations are even in magnetic field.

The lowest-order contribution to the AB current asymmetric in magnetic field is $I^{asymm}_\mu(\phi)=\tilde{I}^e_{\mu\leftarrow\bar{\mu}}+\tilde{I}^h_{\mu\leftarrow\bar{\mu}}-\tilde{I}^e_{\bar{\mu}\leftarrow\mu}-\tilde{I}^h_{\bar{\mu}\leftarrow\mu}$, where $(\mu=L,R=\pm1)$

$$\tilde{I}^{e}_{\mu\leftarrow\bar{\mu}} = \frac{e}{2\pi\hbar}\mu \sin\phi(2\pi)^3 N_\mu (N_{\bar{\mu}})^2 \sum_{\alpha,\alpha',\alpha'',\beta'} P_{\beta'} \int d\epsilon' f_{\bar{\mu}}(\epsilon') \times \Bigg\{ \sum_{\beta} [1-f_\mu(\epsilon'+\epsilon_{\beta'}-\epsilon_\beta)][1-f_{\bar{\mu}}(\epsilon'+\epsilon_{\beta'}-\epsilon_\beta)] V^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'} W_{\mu\alpha;\bar{\mu}\alpha''} V^{\beta;\beta'}_{\bar{\mu}\alpha'';\bar{\mu}\alpha'} - \sum_{\beta''}[1-f_\mu(\epsilon')] \times [1-f_{\bar{\mu}}(\epsilon'+\epsilon_{\beta'}-\epsilon_{\beta''})] W_{\mu\alpha;\bar{\mu}\alpha'} V^{\beta';\beta''}_{\mu\alpha;\bar{\mu}\alpha''} V^{\beta'';\beta'}_{\bar{\mu}\alpha'';\bar{\mu}\alpha'} \Bigg\}, \tag{A10a}$$

$$\tilde{I}^{h}_{\mu\leftarrow\bar{\mu}} = -\frac{e}{2\pi\hbar}\mu \sin\phi(2\pi)^3 (N_\mu)^2 N_{\bar{\mu}} \sum_{\alpha,\alpha',\alpha'',\beta'} P_{\beta'} \int d\epsilon' f_{\bar{\mu}}(\epsilon') \times \Bigg\{ \sum_{\beta} [1-f_\mu(\epsilon'+\epsilon_{\beta'}-\epsilon_\beta)] f_\mu(\epsilon') V^{\beta;\beta'}_{\mu\alpha;\bar{\mu}\alpha'} W_{\mu\alpha'';\bar{\mu}\alpha'} V^{\beta;\beta'}_{\mu\alpha;\mu\alpha'} - \sum_{\beta''}[1-f_\mu(\epsilon')] \times f_\mu(\epsilon'+\epsilon_{\beta''}-\epsilon_{\beta'}) W_{\mu\alpha;\bar{\mu}\alpha'} V^{\beta';\beta''}_{\mu\alpha'';\bar{\mu}\alpha'} V^{\beta'';\beta'}_{\mu\alpha;\mu\alpha''} \Bigg\}. \tag{A10b}$$

The asymmetric nature of these terms is evident from their proportionality to the sine, $\sin\phi$, of the magnetic flux.

The first and the second terms in the curly brackets in Eqs. (A10) describe, respectively, inelastic and elastic processes. In the case when $\beta=\beta'=\beta''$ the two terms cancel out, i.e., the only terms that contribute are those that involve a change in the QD state [types (ii) and (iii) processes in the discussion above]. We intentionally kept the order of the matrix elements from Eqs. (A5), so one can readily see that the only processes that give nonzero contribution to the asymmetric current are those that begin with the creation of an electron-hole pair in one of the leads (matrix element $V^{\beta;\beta'}_{\bar{\mu}\alpha'';\bar{\mu}\alpha'}$ or $V^{\beta;\beta'}_{\mu\alpha;\mu\alpha'}$ with $\beta\neq\beta'$).

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[14] We assume single-channel leads and include the magnetic field explicitly only via Aharonov-Bohm phase. A more general analysis can be performed based on the properties of the tunneling matrix elements in respect to reversal of magnetic field, see Secs. 1 and 2 of Appendix.

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