Breaking of Phase Symmetry in Non-Equilibrium Aharonov-Bohm Oscillations through a Quantum Dot

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(Dated: February 16, 2009)

Linear response conductance of a two terminal Aharonov-Bohm (AB) interferometer is an even function of magnetic field. This phase symmetry is no expected to hold beyond the linear response regime. 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 letter we discuss the processes that break AB phase symmetry. In particular we 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 of the AB phase. We show that our theoretical results are consistent with experimental findings.

PACS numbers: 73.23.-b, 73.23.Hk, 73.63.Kv

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 \textsuperscript{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 \(\pi\) (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 \textsuperscript{3} and is, in fact, a manifestation of more general linear-response Oulser-Büttiker symmetries \textsuperscript{4,5}. Deviations from phase symmetry in two-terminal devices in the nonlinear regime have been studied theoretically \textsuperscript{6,7,8}, as well as in experiments on AB cavities \textsuperscript{9} and AB rings \textsuperscript{10}. The resulting phase of the AB oscillations changes smoothly (almost linearly) with increasing voltage bias \textsuperscript{10}.

Rather puzzlingly, a recent experiment \textsuperscript{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 \(V\) in a highly non-monotonous fashion: it remained close 0 and \(\pi\), 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 \(\Delta\), 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, presence of the finite bias threshold for the inelastic cotunneling renders inapplicable the methods based on expansion in powers of the bias voltage \textsuperscript{11}, and thus cannot explain the experimental observations. In this Letter 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. Based on their dependence on voltage bias and magnetic field we establish that the bias dependence of the AB phase is highly non-monotonous. In particular, (i) the oscillations indeed remain symmetric up to the onset of inelastic cotunneling (\(eV \simeq \Delta\)) (i.e. with AB phase 0 or \(\pi\)), in agreement with experiments; (ii) with onset of inelastic cotunneling, AB oscillations acquire non-zero 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) \textsuperscript{12}. The theoretical findings are supported by the in-depth analysis of the experimental data of Ref. \textsuperscript{11}.

\textbf{Theoretical formulation} We consider an AB interferometer schematically shown in Fig. 1. One arm of the interferometer contains a QD which is assumed to be in 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 \textsuperscript{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 $H_\mu = \sum_E E^\dag_\mu E_\mu$ is the Hamiltonian of electrons in lead $\mu = L, R$. $E$ labels energy states within one lead. $H_D = \sum_\beta \epsilon_\beta d^\dag_\beta d_\beta$ is the Hamiltonian of the QD, which contains only one electron and has energy levels $\epsilon_\beta$. $c_{\mu E}$ destroys a lead electron in state $\mu E$, $d_\beta$ destroys QD state $\beta$.

$W$ 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 change of the QD state. These terms in the Hamiltonian are given by

$$W = \sum_{\mu E} \sum_{\mu' E'} W^\dag_{\mu' \mu E'} e_{\mu' E'}^\dag c_{\mu E}$$ (1a)

$$V = \sum_{\beta, \beta'} \sum_{\mu E} V_{\beta \beta'}^\dag_{\mu E} d_{\beta'}^\dag c_{\mu E} e_{\beta'}$$ (1b)

where $W_{\mu' \mu E'}$ and $V_{\beta \beta'}^\dag_{\mu E}$ are real, and $\phi$ is the magnetic flux through the interferometer ($\phi_{RL} = -\phi_{LR} = \phi$, $\phi_{LL} = \phi_{RR} = 0$).

**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 (where $\epsilon_0$ 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 Fig. 2. a, b, are, respectively,

$$4\pi \Re \left[ \frac{(W_{R,L}e^{i\phi})^* V_{R,R}^{1,2} V_{R,L}^{2,1}}{\epsilon_1 + E_L - \epsilon_R - i0^+} \right] \delta(E_L - E_R)$$ (2a)

$$4\pi \Re \left[ \frac{V_{R,L}^{2,1} W_{R,L} e^{i\phi}}{\epsilon_1 + E_L - E_R + i0^+} \right] \delta(E_L + \epsilon_1 - \tilde{E}_R - \epsilon_2)$$ (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 correction to the current, the probabilities in Eq. (2) are multiplied by the factor $P_{1fL(E_L)}[1 - f_R(E_R)][1 - f_R(\tilde{E}_R)]$ (which also limits possible intermediate states) and integrated over $E_L, E_R$ and $\tilde{E}_R$.

The contribution to AB oscillations coming from the real parts of the denominators in Eqs. (2) is even in magnetic field. Asymmetric terms may result 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 the contribution to AB oscillations odd in magnetic field may result only from the 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 $E_L + \epsilon_1 = E_R + \epsilon_1 = \tilde{E}_R + \epsilon_2$. This is the case shown in all our figures.

The asymmetric contribution due to the process (2)a is thus given by

$$-2W_{R,L} V_{R,R}^{1,2} V_{R,L}^{2,1} \delta(\epsilon_1 + E_L - \epsilon_2 - \tilde{E}_R) \delta(E_L - E_R) \sin \phi$$ (3)

On the other hand, the asymmetric contribution of the process (2)b is given by exactly the same expression, but with the opposite sign, and thus the asymmetric contri-
bution is canceled between these two processes. This is no surprise. The first process (Fig. 2a) 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, and 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. 2b) 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, Fig. 2c, d.

However, let us examine the process shown in Fig. 2e. The process that should cancel its asymmetric contribution is depicted in Fig. 2e. 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. 2e gives rise to AB oscillations odd in magnetic field. Similar cancellation occurs for the processes starting with the QD in its exited state, Fig. 2c, d.

Overall, this means that the phase of AB oscillations is not a monotonous function of bias: it is usually very close to 0, π, but deviates significantly from these values when phase switching occurs.

**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. In order to judge whether oscillations are strictly symmetric or not we provide in the lower left panel of Fig. 3 the colorplot for 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 colorplots 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 $V = \pm 2.5 V$ 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 non-zero 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

$$P(Vsd) = \sqrt{B_{max} dG_{asym}^2 (B, Vsd) / (B_{max} - B_{min})},$$

where $G_{asym} (B, VSD)$ is the asymmetric component of the differential conductance as a function of magnetic field $B$ and bias voltage, $V_{sd}$. 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 of 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 non-zero 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].

Although the asymmetric component makes the AB
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 order in lead-to-lead tunneling than the symmetric one, the AB phase remains close to values 0 and $\pi$. 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 $\pi$. We show that our theoretical findings are in excellent agreement with the experimental data of Ref. [11].

We thank Y. Gefen, V. Kashcheyevs, T. Aono and M. Khodas for useful discussions. We are grateful to O. Entin-Wohlman and A. Golub for valuable comments. This work was supported in part by the ISF and BSF. V.P. is partially supported by Pratt Fellowship.

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