Kondo-induced electric polarization modulated by magnetic flux through a triangular triple quantum dot

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Abstract. The Kondo effect plays an important role in emergence of electric polarization in a triangular triple-quantum-dot system, where one of the three dots is point-contacted with a single lead, and a magnetic flux penetrates through the triangular loop. The Kondo-induced electric polarization exhibits an Aharonov-Bohm type oscillation as a function of the magnetic flux. Our theoretical study shows various oscillation patterns associated with the field-dependent mixing of twofold orbitally degenerate ground states and their sensitivity to the point contact.

1. Introduction
Recent nanofabrication techniques have accelerated investigation of the Kondo effect in various multiple-quantum-dot systems [1, 2, 3]. The quantum dots connected to metallic leads are considered as artificial variants of magnetic impurities embedded in bulk materials [4, 5, 6]. A geometric configuration of quantum dots is a new aspect in the Kondo physics. A triangular-triple-quantum-dot (TTQD) system is an ideal example that exhibits a cross correlation between spin and charge degrees of freedom associated with the Kondo effect [7, 8]. In our previous studies [9, 10], we showed the emergence of electric polarization in the TTQD point-contacted with a single lead, accompanied by the spin reconfiguration. This magnetoelectric effect was originally proposed for an emergent electric polarization in bulk systems such as spin-frustrated Mott insulators with triangular unit cells [11, 12, 13]. It must be distinguished from the spin-orbit-coupling mediated mechanisms widely argued for multiferroic properties [14, 15].

In this paper, we demonstrate a magnetic flux effect on the Kondo-induced electric polarization, which is experimentally controllable by fine-tuning a point contact in the TTQD system. The electron paths in the triangular loop can be regarded as molecular orbitals. Since the coherency of the electronic state in the loop causes interference phenomena, it is expected that the electric polarization exhibits a periodic oscillation as a function of the magnetic flux penetrating through the loop. This Aharonov-Bohm (AB) type oscillation is sensitive to the strength of the point contact with the lead [16]. We elucidate that the electric polarization displays various oscillation patterns as a consequence of field-dependent mixing of the different-parity orbital states of TTQD.
2. Model

For the TTQD states, we focus on a half-filled case in the strong Coulomb coupling limit, using a three-site Hubbard model with a triangular loop shown in Fig. 1(a). The orbital states of TTQD are modulated by the magnetic flux $\Phi$ penetrating through the loop. To investigate the Kondo effect in the TTQD with a single lead, we consider the following Anderson model:

$$H = H_{\text{lead}} + H_{\text{dots}} + H_{\text{mix}},$$

$$H_{\text{lead}} = \sum_{\kappa \sigma} \varepsilon_{k} c_{\kappa \sigma}^\dagger c_{\kappa \sigma},$$

$$H_{\text{dots}} = -\sum_{i \neq j} t_{ij} \sum_{\sigma} d_{i \sigma}^\dagger d_{j \sigma} + \sum_{i} \left( \varepsilon_{d,i} + \frac{U}{2} \right) n_{i} + \frac{U}{2} \sum_{i} (n_{i} - 1)^{2} \ (i, j = a, b, c),$$

$$H_{\text{mix}} = \sum_{\kappa \sigma} \left( v_{k} d_{i \sigma}^\dagger c_{\kappa \sigma} + v_{k} c_{\kappa \sigma}^\dagger d_{i \sigma} \right),$$

where $H_{\text{lead}}$ is the Hamiltonian for the kinetic energy $\varepsilon_{k}$ of the lead electrons with the wave vector $k$. $H_{\text{dots}}$ is for the energy of the TTQD electrons hopping around the three sites (labelled $a$, $b$, and $c$). The last term $H_{\text{mix}}$ represents electron transfer through the point contact of the lead connected to the only $a$ site. The creation and annihilation operators $c_{\kappa \sigma}$ and $c_{\kappa \sigma}^\dagger$ ($d_{i \sigma}$ and $d_{i \sigma}^\dagger$) are for the lead electrons (the $i$-th site electron) with spin $\sigma = \uparrow, \downarrow$. In Eq. (3), $t_{ij}$, $\varepsilon_{d,i}$ and $U$ are the intersite electron hopping matrix element, the $i$-th site intradot orbital energy and the on-site Coulomb coupling, respectively. In Eq. (4), the hybridization strength $v_{k}$ is related to the level broadening of $\Gamma \equiv \pi \rho |v_{k}|^{2}$ ($\rho$ is the density of states of the conduction electrons at the Fermi energy), where the $k$ dependence is neglected. For simplicity, we only consider the magnetic flux effect on the TTQD loop, and neglect any magnetic-field effect on the electron spins, e.g. the Zeeman effect. This is valid for a weak magnetic field that does not destroy the Kondo-Singlet formation at the $a$ site. Throughout the paper, we assume that the three sites of TTQD are equivalent, and each site satisfies the symmetric condition $\varepsilon_{d,i} = -U/2 < 0$ [17]. In this equilateral TTQD case, the magnetic flux effect is considered by introducing additional phase factors in the electron hopping matrices as $t_{ij} = t e^{i\varphi_{ij}}$ ($t > 0$), where $\varphi_{ab} = \varphi_{bc} = \varphi_{ca} = \varphi/3$ ($\varphi_{ij} = -\varphi_{ji}$) [18, 19]. The phase $\varphi = 2 \pi \Phi/\Phi_{0}$ is measured by the unit of magnetic flux quantum $\Phi_{0} = hc/e$ ($h$ is the Planck constant, $c$ is the speed of light in vacuum, and $e$ is the elementary charge).

Figure 1(b) shows the emergence of electric polarization by the Kondo effect at low temperatures, which is related to the following orbitally degenerate ground states of the isolated TTQD:

$$|\phi_{g+}\rangle = \frac{1}{\sqrt{2}} d_{a \uparrow}^\dagger (d_{b \uparrow}^\dagger d_{c \downarrow} - d_{b \downarrow}^\dagger d_{c \uparrow})|0\rangle,$$

$$|\phi_{g-}\rangle = \frac{1}{\sqrt{6}} [d_{a \uparrow}^\dagger (d_{b \uparrow}^\dagger d_{c \downarrow} + d_{b \downarrow}^\dagger d_{c \uparrow}) - 2d_{a \downarrow}^\dagger d_{b \uparrow}^\dagger d_{c \uparrow}]|0\rangle,$$

for the $S_{z} = 1/2$ spin states, where $|0\rangle$ is a vacuum state. The time reversal $S_{z} = -1/2$ states are given by interchanging the spin-up and spin-down subscripts of $d_{i \sigma}^\dagger$. The orbitals are denoted by even ($+$) and odd ($-$) parities against interchange of the $b$ and $c$ sites. Therefore, the TTQD ground states are fourfold degenerate with respect to the spin and orbital degrees of freedom when the magnetic flux is absent ($\Phi = 0$). For a finite $\Phi$, both even and odd parities are mixed as $|\phi_{g+}\rangle \pm i|\phi_{g-}\rangle$.

To examine the Anderson model Hamiltonian in Eq. (1), we use the numerical renormalization group (NRG) method [20, 21]. For $\Phi \neq 0$, it is convenient to introduce new local-electron operators $d_{c \sigma} = (d_{b \sigma} + d_{c \sigma})/\sqrt{2}$ and $d_{c \sigma} = i(d_{b \sigma} - d_{c \sigma})/\sqrt{2}$ for the even- and odd-parity orbitals,
respectively, between the $b$ and $c$ sites. In terms of the three $a$, $e$ and $o$ orbital bases, the NRG Hamiltonian is given as a symmetric matrix form appropriate to the numerical calculation [16]. We use $\Lambda = 3$ for the logarithmic discretization parameter for conduction electrons and retain about 2000 low-lying states at each renormalization step. The half width of the conduction band is chosen as the unit of energy and temperature, and the Coulomb coupling is fixed at $U = 0.9$ throughout the paper.

3. Results

3.1. Origin of electric polarization

For the TTQD loop, the charge distribution is closely related to the spin configuration as follows [9]. We define the electric polarization as $\delta n = (2n_a - n_b - n_c)/3$, where $n_i$ is the electron occupation number at the $i$-th site. According to Bulaevskii et al. [11], the operator for the electric polarization is expressed by the correlation among on-site spin operators $S_i$ ($i = a, b, c$) as

$$\delta \hat{n} = 8 \text{Re} \left( \frac{t_{ab} t_{bc} t_{ca}}{U^3} \right) \left[ S_a \cdot (S_b + S_c) - 2S_b \cdot S_c \right],$$

as a consequence of electron hopping in the closed loop. This indicates that no electric polarization emerges when all spins are identical. In the TTQD Kondo system presented here, the local spin $S_a$ is compensated for by the conduction electrons at low temperatures. As a result, the electric polarization is induced in accordance with

$$\delta n = -16 \left( \frac{t}{U} \right)^3 \cos \varphi \cdot \langle S_b \cdot S_c \rangle,$$

where the $\cos \varphi$ behavior comes from $t_{ab} t_{bc} t_{ca} = t^3 e^{i\varphi}$. The expectation value $\langle S_b \cdot S_c \rangle$ is given by the mixing of the $b$-$c$ spin-singlet and spin-triplet states, which is related to the $\varphi$-dependent mixing between $|\phi_{g+}\rangle$ and $|\phi_{g-}\rangle$. This is the essence of the Kondo-induced electric polarization. It differs from a simple symmetry-lowering effect such as the deformation of the TTQD geometry.
3.2. Temperature dependence of oscillating electric polarization

Figure 2 shows the electric polarization $\delta n$ with a periodic oscillation as a function of $x = \varphi/\pi$ at various temperatures $T$. Each data is given in the range $0 \leq x \leq 2$. $\delta n$ is always zero irrespective of $T$ at $x = \pm 0.5, \pm 1.5, \cdots$, since the particle-hole symmetry is conserved there. These nodes are also related to $\cos \varphi = 0$ in Eq. (8). At high $T$, $\delta n$ exhibits a cosine curve with a small amplitude, owing to the small energy splitting of $|\phi_{g\pm}\rangle$ by the point contact at the $a$ site. This symmetry-lowering effect is not important practically, since the Kondo effect and the TTQD intersite spin interaction compete with each other, which results in almost degenerate $|\phi_{g\pm}\rangle$ in the high temperature region. Therefore, $\delta n \approx 0$ at high $T$ is due to the cancellation of the opposite contributions from $|\phi_{g\pm}\rangle$, namely, $\langle \phi_{g\pm} | \delta \hat{n} | \phi_{g\pm} \rangle = \pm 12 (t/U)^3 \cos \varphi$.

The Kondo-induced electric polarization at $x = 0, \pm 1, \pm 2, \cdots$ is explained as follows [9]. In Fig. 2, the abrupt increase of $|\delta n|$ at the intermediate $T$ is caused by the Kondo-singlet formation at the $a$ site. At sufficiently low temperatures, $\delta n$ approaches a saturation value $\delta n^* \approx \pm 12 (t/U)^3$ for $\Gamma/U \ll 1$. In the TTQD, the $b$-$c$ dimerized spin-singlet is decoupled from the Kondo singlet, which means that the Kondo effect stabilizes $|\phi_{g+}\rangle$ against $|\phi_{g-}\rangle$. This is also confirmed by Eq. (8). The electric polarization $\delta n^*$ is prominent at low temperatures as a consequence of $\langle S_b \cdot S_c \rangle \approx -3/4$ for the $|\phi_{g+}\rangle$ dominant ground state.

It is also important that $\delta n$ depends on the three electron hopping parameters $t_{ij}$ including their phase shifts $\varphi_{ij}$ in the TTQD loop. This interference effect is usually independent of the TTQD spin configuration. Notice that $\delta n(\varphi)$ deviates from the $\cos \varphi$ behavior at low $T$ in Fig. 2, which is the most pronounced feature of the Kondo-induced electric polarization. While $|\phi_{g+}\rangle$ is stabilized by the Kondo effect, both $|\phi_{g\pm}\rangle$ states are mixed through the magnetic flux effect. As a result, $|\phi_{g+}\rangle$ participates in the Kondo singlet as well as $|\phi_{g-}\rangle$, leading to the TTQD spin reconfiguration. It is predicted by Eq. (8) that the expectation value $\langle S_b \cdot S_c \rangle$ changes from $-3/4$ to $0$ in the ground state. For $\langle S_b \cdot S_c \rangle = 0$, the TTQD ground state is given as $|\phi_{g+}\rangle \pm i|\phi_{g-}\rangle$, corresponding to a clockwise or counterclockwise orbital state of TTQD. Owing to the Kondo effect, the $\varphi$ dependent mixing of $|\phi_{g\pm}\rangle$ is very sensitive to the strength $\Gamma$ of the point contact, as discussed below.

3.3. AB type oscillation controlled by Kondo effect

Figure 3 shows $\delta n^*$ in the low temperature limit for the Kondo couplings $\Gamma/U = 0.0473$(A), 0.0946(B) and 0.141(C). The data are plotted in the half period $0 \leq x \leq 1$, where (B) corresponds to $\delta n$ at the lowest $T$ (black curve) in Fig. 2. The data (A) for the smallest $\Gamma$ exhibits sharp peaks at $x = 0, 1$. As $\Gamma$ decreases further, the peaks become more singular (not shown). In the $\Gamma \to 0$ limit, $\delta n^*$ vanishes except at $x = 0, \pm 1, \pm 2, \cdots$. This is because the TTQD ground state is given by the equal weight mixing of even- and odd-parity states as $|\phi_{g+}\rangle \pm i|\phi_{g-}\rangle$. At the singularities, on the other hand, the Kondo effect hinders the $|\phi_{g\pm}\rangle$ mixing, leading to the $|\phi_{g+}\rangle$ dominant state in the Kondo-singlet formation. Thus, the spikelike behavior of (A) indicates a clear evidence of the competition between the Kondo and magnetic flux effects. For a larger $\Gamma$, $|\phi_{g+}\rangle$ becomes more dominant in the TTQD ground state at low temperatures, and the $\delta n^*$ peaks become broader. As a result, $\delta n^*(x)$ shows a cosinelike curve as (C). In addition, the amplitude of the $\delta n^*$ oscillation is considerably reduced by the strong Kondo coupling. Therefore, for a large $\Gamma$, the charge distribution becomes almost uniform over the three sites of TTQD.

Next, we discuss quantitatively the $\Gamma$ dependence of the $\delta n^*$ oscillation. It is convenient to express $\delta n^* = \delta n_0^* f_\varphi \cos \varphi$ by means of the $\varphi = 0$ value $\delta n_0^* \sim (t/U)^3$. In terms of the $\varphi$ dependent function $f_\varphi$, the $\delta n^*$ oscillation changes from the spikelike behavior to the cosinelike curve with increasing $\Gamma/U$. When the TTQD ground-state wave function can be approximated by $\alpha_+|\phi_{g+}\rangle + \alpha_-i|\phi_{g-}\rangle$, a simple expression of $f_\varphi$ is given as $f_\varphi = 1 - \alpha_-^2/\alpha_+^2$. As discussed in Appendix, the $\Gamma$ dependence of $f_\varphi$ can be reproduced by the symmetry-lowering parameter $\kappa$. Indeed, $f_\varphi$ changes from 0 to 1 continuously as $\Gamma$ increases in the weak Kondo coupling case.
Concerning the experimental controllability with the point contact, we have to consider the characteristic temperature for the emergence of electric polarization which is lowered by decreasing $\Gamma$. The spikelike peaks for a weak point contact might be difficult to detect since they only become prominent at relatively low temperatures. On the other hand, a strong point contact considerably reduces the amplitude of the oscillation. Thus, it is important to optimize...
Γ for the practical observation of the Kondo-induced polarization.

Appendix: Mixing of different-parity orbital states
Here, we show how the ϕ-dependent mixing of |ϕg±⟩ is affected by the Kondo effect at low temperatures, which resembles a symmetry-lowering effect on the isolated TQQD as follows [16]. In Eq. (3), we introduce a symmetry-lowering parameter κ as |tab⟩ = |tca⟩ = κt (0 < κ < 1) and |tbc⟩ = t. The perturbation theory is applied to the Schrödinger equation for H dots, and the eigenvalue problem is solved by a series expansion up to the third order of ̃t (≡ t/U). When the ground-state wave function is expressed by |ψ⟩ = α+|ϕg+⟩ + α−i|ϕg−⟩ (the coefficients α± are real), we obtain

\[
\begin{align*}
U ̃t^2 \begin{pmatrix}
-4 - 2κ^2 & (-6\sqrt{3}κ^2 \sin \varphi) ̃t \\
(-6\sqrt{3}κ^2 \sin \varphi) ̃t & -6κ^2
\end{pmatrix}
\begin{pmatrix}
α_+ \\
α_-
\end{pmatrix}
= E \begin{pmatrix}
α_+ \\
α_-
\end{pmatrix},
\end{align*}
\]

where E is the eigenvalue. This equation indicates |α+| ≫ |α−| for κ ≪ 1 corresponding to the |ϕg+⟩ dominant ground state for a large Γ/U. The |α+| ≫ |α−| solution also holds for a sufficiently small |sin ϕ| even if κ is not so small (more precisely, |sin ϕ| < 0.2(κ−2 − 1)/̃t). This is related to the periodic peaks of δnκ* that become prominent for a small Γ. On the other hand, |α+| ≈ |α−| is obtained for κ ≈ 1 and |sin ϕ| ≈ 1, which corresponds to the equal weight mixing of |ϕg±⟩ for an extremely small Γ/U. The electric polarization is calculated as

\[
\langle ψ | δn | ψ \rangle = 12κ^2 \cos \varphi \cdot (α_+^2 - α_-^2)̃t^3.
\]

Notice that it vanishes in the isolated TQQD with equilateral triangular symmetry (κ = 1), where |α+| = |α−| holds for the degenerate |ϕg±⟩ states.

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