Adiabatic Pumping in the Mixed-Valence and Kondo Regimes

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We investigate adiabatic pumping through a quantum dot with a single level in the mixed-valence and Kondo regimes using the slave boson mean field approximation. The pumped current is driven by a gauge potential due to time-dependent tunneling barriers as well as by the modulation of the Friedel phase. The sign of the former contribution depends on the strength of the Coulomb interaction. Under finite magnetic fields, the separation of the spin and charge currents peculiar to the Kondo effect occurs.

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Introduction.—Adiabatic pumping in a quantum dot system occurs when the dot is under slowly varying external gate voltages with zero bias voltage. After a certain period when the system returns to its initial state, a finite charge is transferred by electron interference through the system. In recent experiments, both charge [1] and spin pumping [2] are realized in open quantum dot systems. This pumping has been investigated theoretically; charge pumping [3–11] as well as spin pumping [12–18]. It has been elucidated that the pumping can be understood in terms of the Berry phase argument in Ref. [17] and Refs. [19–22].

Adiabatic pumping is investigated for the systems under electron-electron interactions [12,14,23–26]. In quantum dots, the interactions introduce a prominent feature, the Kondo effect [27–30], where the spin exchange between electrons in the leads and the dot results in an enhancement of the conductance at low temperatures. We investigate the adiabatic pumping in the mixed-valence and the Kondo regimes to demonstrate an interplay between the Kondo effect and a gauge potential associated with the Berry phase, though this system itself is partly studied in Ref. [26]. To this end, we will show explicitly the appearance of the Berry phase term due to time-dependent tunneling barriers and elucidate the connection with the adiabatic pumping under the electron correlations. We will then show that the spin-charge separation peculiar to the Kondo effect emerges as the separation of pumped spin and charge under a finite magnetic field.

Model.—We consider a system which consists of a quantum dot which has a single energy level and couples to two leads, described by the Anderson model [27,28]: $H = \sum_{k,\sigma\alpha=L,R}[\epsilon_k - \mu(t)]c^\dagger_{k\sigma\alpha}c_{k\sigma\alpha} + \sum_{k,\sigma\alpha=L,R}V_\alpha(t)\times(c^\dagger_{k\sigma\alpha}d_\sigma + \text{H.c.}) + H_{\text{dot}}$ with $H_{\text{dot}} = \sum_{\sigma=\pm}E_0 d^\dagger_\sigma d_\sigma + un_+n_-$. Here $c^\dagger_{k\sigma\alpha}$ creates an electron with energy ϵ_k and spin σ in lead $\alpha = L, R$. $d^\dagger_\sigma$ creates an electron in the dot with spin σ, $n_\sigma = d^\dagger_\sigma d_\sigma$, and u is the strength of the Coulomb interaction. We have introduced time-dependent sources, a common chemical potential $\mu(t)$, and tunneling barriers $V_\alpha(t)$ which are driving forces for electron pumping.

Let us first find out a source-drain voltage embedded in the model. To this end, we apply the following unitary transformation for electrons in the leads:

$$\begin{pmatrix} c_{k\sigma+} \\ c_{k\sigma-} \end{pmatrix} = \begin{pmatrix} \xi & \eta \\ -\eta & \xi \end{pmatrix}\begin{pmatrix} c_{k\sigma L} \\ c_{k\sigma R} \end{pmatrix} \equiv U^{-1}\begin{pmatrix} c_{k\sigma L} \\ c_{k\sigma R} \end{pmatrix} \quad (1)$$

with $\xi = V_L/V$, $\eta = V_R/V$, and $V = \sqrt{|V_L|^2 + |V_R|^2}$ [27]. If V_α are constant, the Hamiltonian reduces to the single impurity Anderson model; The modes $c_{k\sigma-}$ decouple with electrons in the dot while the modes $c_{k\sigma+}$ couple with them through the tunneling matrix element V.

When V_α are time dependent, the unitary transformation generates the Berry phase term $(c^\dagger_{k\sigma+}, c^\dagger_{k\sigma-})[-i\hbar U^{-1}(t)\partial U(t)/\partial t](c_{k\sigma+}, c_{k\sigma-})^t = -\sum_{k\sigma}[c^\dagger_{k\sigma+}i\hbar a(t)c_{k\sigma-} - c^\dagger_{k\sigma-}i\hbar a(t)c_{k\sigma+}]$ with a gauge potential $a(t) = (-\xi\dot{\eta} + \dot{\xi}\eta)$; the modes $c_{k\sigma+/-}$ are mixed. To diagonalize the Hamiltonian again, we apply an additional time-*independent* unitary transformation $b_{k\sigma s} = 1/\sqrt{2}(c_{k\sigma+} + isc_{k\sigma-})$ $(s = \pm)$ resulting in

$$H = \sum_{k,\sigma,s=\pm} b^\dagger_{k\sigma s}[\epsilon_k - (\mu(t) + s\hbar a(t))]b_{k\sigma s} + \frac{V(t)}{\sqrt{2}}\sum_{k,\sigma,s=\pm}[b^\dagger_{k\sigma s}d_\sigma + \text{H.c.}] + H_{\text{dot}}. \quad (2)$$

Equation (2) defines that the gauge potential $a(t)$, which originates from the time-dependent barriers, acts as a finite bias voltage between $b_{k\sigma+}$ and $b_{k\sigma-}$ fields. Hence a finite current flows through the dot even when a real bias voltage is zero. In the following, we assume $u = 0$ for a while.

Adiabatic current and pumped charge.—The current $I_\sigma = \langle \hat{I}_{\sigma,L} - \hat{I}_{\sigma,R}\rangle/2$ with $\hat{I}_{\sigma,\alpha} = ie/\hbar\sum_k V_\alpha(t)(d^\dagger_\sigma c_{k\sigma\alpha} - c^\dagger_{k\sigma\alpha}d_\sigma)$ can be represented by $b^\dagger_{k\sigma s}$ fields:

$$I_\sigma = \frac{ieV(t)}{2\sqrt{2}\hbar}\sum_{\substack{k \\ s=\pm}}[(\xi + is\eta)^2\langle d^\dagger_\sigma b_{k\sigma s}\rangle - \text{C.c.}]. \quad (3)$$

We consider the adiabatic regime where the frequency Ω of external time-dependent sources is much smaller than the dot-lead coupling $\Gamma(t) = \pi\rho|V(t)|^2$, with the density

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of states ρ at the Fermi energy in the leads: $\Omega \ll \Gamma(t)/\hbar$. This condition is consistent with the one discussed in Ref. [10]. In this limit, Eq. (3) can be written as $I_\sigma = I_{\sigma,B} + I_{\sigma,F}$ with

$$I_{\sigma,B}(t) = -\frac{2e\Gamma(t)\xi(t)\eta(t)}{2\pi}a(t) \times \int d\epsilon[G^R_\sigma(\epsilon;t) + G^A_\sigma(\epsilon;t)]\left(-\frac{\partial f}{\partial \epsilon}\right), \qquad (4)$$

and

$$I_{\sigma,F}(t) = e\frac{[\xi^2(t) - \eta^2(t)]}{2}\frac{dn_{\sigma;\mathrm{dot}}}{dt}, \qquad (5)$$

where $G^{R/A}(\epsilon, t) = 1/[\epsilon - E_0 \pm i\Gamma(t)]$ are the retarded (advanced) Green functions of electrons in the dot, f is the Fermi-Dirac function, and $n_{\sigma;\mathrm{dot}}$ is the occupation number in the dot. (The derivation will be discussed later.) We emphasize that $I_{\sigma,B}$ is proportional to the real part of the dot Green function G^R; in contrast the conductance given by the Landauer formula is proportional to the imaginary part. This difference indicates that the adiabatic pumping will convey additional information on the electronic state in the dot, which is not captured by the conductance. We further notice that $I_{\sigma,F}$ always accompanies the change of $n_{\sigma;\mathrm{dot}}$ when it flows while $I_{\sigma,B}$ does not. Note that similar expressions are obtained in a recent paper [11] using a different formalism [9]. For simplicity, we consider zero temperature limit in the following.

Equations. (4) and (5) are also derived by the conventional scattering matrix approach of the pumping [3,6,31]. The pumped charge Q_σ per cycle is

$$Q_\sigma = \frac{e}{2\pi}\oint dt(1 - T_\sigma)\frac{d\alpha_\sigma}{dt} \equiv \oint dt I_{\sigma;S}(t) \qquad (6)$$

with the transmission probability T_σ and the phase of the reflection coefficient α_σ through the dot. The scattering matrix of the dot is given by

$$U\begin{pmatrix} e^{2i\delta_\sigma} & 0 \\ 0 & 0 \end{pmatrix}U^{-1} = \begin{pmatrix} \xi^2 e^{2i\delta_\sigma} + \eta^2 & \xi\eta(e^{2i\delta_\sigma} - 1) \\ \xi\eta(e^{2i\delta_\sigma} - 1) & \xi^2 + \eta^2 e^{2i\delta_\sigma} \end{pmatrix} \qquad (7)$$

with the Friedel phase δ_σ at the Fermi energy in the leads. It determines $T_\sigma = 4\xi^2\eta^2\sin^2\delta_\sigma$, and $\alpha_\sigma = \arctan[(\xi^2 - \eta^2)\tan\delta_\sigma]$. Hence,

$$I_{\sigma;S} = \frac{e}{2\pi}2\xi\eta a\sin 2\delta_\sigma + \frac{e}{2\pi}(\xi^2 - \eta^2)\frac{d\delta_\sigma}{dt}. \qquad (8)$$

The first term corresponds to Eq. (4) and the second to Eq. (5). These are due to (i) the relation of $\exp(2i\delta_\sigma) = 1 - 2i\Gamma G^R(\epsilon = 0; t)$ and (ii) the Friedel sum rule [32]: $n_{\sigma;\mathrm{dot}} = \delta_\sigma/\pi$. (See the discussion later.) Pumped charge Q_σ is then represented by two parameters, δ_σ and θ, which defines $\xi = \cos\theta$ and $\eta = \sin\theta$ ($0 \leq \theta \leq \pi/2$):

$$Q_\sigma = \frac{e}{2\pi}\oint(\cos 2\theta d\delta_\sigma - \sin 2\theta \sin 2\delta_\sigma d\theta). \qquad (9)$$

The Friedel phase δ_σ will be controlled by the gate voltage $V_g = E_0 - \mu$. Furthermore, δ_σ is independent of θ since the effect of θ is only involved in the Hamiltonian (2) as the factor $a(t) = -d\theta/dt$, which is small in the adiabatic limit and can be disregarded. Thereby, $\delta_\sigma = \delta_\sigma(V_g)$. In the following, we choose V_g and θ as the pumping parameters (see Fig. 1).

Pumping under the Coulomb interaction.—Now we investigate the effect of u in the mixed-valence and Kondo regimes. To this end, we adopt the slave boson mean field approximation [33,34], which has been successfully applied for the regimes. We assume $u \to \infty$ to exclude the double occupancy of electrons in the dot. In this situation, the annihilation operator d_σ of electron in the dot is written as $d_\sigma = b^\dagger f_\sigma$ with the slave boson operator b and the pseudo fermion operator f_σ with the constraint term of $H_c = \lambda(\sum_\sigma f^\dagger_\sigma f_\sigma + b^\dagger b - 1)$, where λ is a Lagrange multiplier. We assume that b and λ are constant, determined by the self-consistent equations as a function of the gate voltage V_g [33,34]. After the approximation, the dot level and dot-lead coupling are renormalized: $\tilde{E}_0 = E_0 + \lambda$, and $\tilde{\Gamma} = \Gamma b^2$ [35]. The current is therefore given by the sum of Eqs. (4) and (5) with these renormalized values. Note that to keep the adiabatic condition, we need a smaller value of Ω; $\Omega \ll \tilde{\Gamma}/\hbar$, since $\tilde{\Gamma} < \Gamma$.

Let us show that $I_{\sigma,B}$ can flow in the opposite direction depending on the strength of u. To this end, we choose the pumping path as shown in Fig. 1(a), where $Q = Q_\sigma = \frac{e}{2\pi}\sin^2\frac{\delta_1 - \delta_2}{2}\sin(\delta_1 + \delta_2)$. (The contribution from $I_{\sigma,F}$ is cancelled.) In Fig. 2, Q (in the unit of $e/2\pi$) is plotted as a function of V_2 for $V_1 = \Gamma$ ($\delta_1 < \delta < \delta_2$). The solid and dashed lines represent the result for the interacting ($u \to \infty$) and noninteracting ($u = 0$) models, respectively. The qualitative difference in Q between the two models appears when the electron interaction is essential ($V_2 < -\Gamma$). When $u = 0$, Q is negative, while when $u \to \infty$ it is positive; the pumped current flows in the opposite directions. If we choose the path as in Fig. 1(b), only the contribution from $I_{\sigma,F}$ remains and $Q = \frac{e}{\pi}(\delta_2 - \delta_1)$.

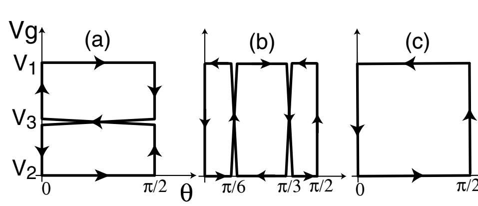

FIG. 1. Pumping paths in the (θ, V_g) plane. V_3 is defined through a relation for δ: $\delta(V_3) = [\delta(V_2) + \delta(V_1)]/2$.

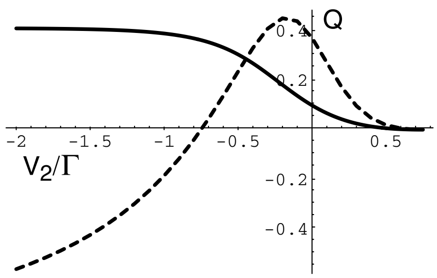

FIG. 2. Pumped charge Q (in the unit of $e/2\pi$) for the path in Fig. 1(a) as a function of V_2 for $V_1 = \Gamma$. The solid and dashed lines stand for the interacting ($u \to \infty$) and noninteracting ($u = 0$) models, respectively.

Then the sign change of Q does not happen. The conductance also does not show such a change of sign.

Spin pumping in the Kondo regime.—Let us look at the Kondo effect under a finite magnetic field, where the Zeeman energy E_Z lifts spin degeneracy of the dot level: $E_0 \to E_0 \pm E_Z$. In Fig. 3(a), the Friedel phases $\delta_\pm$ are plotted as a function of the gate voltage V_g under $E_Z = 0.5 \times 10^{-3}\Gamma$. When $V_g < -\Gamma$, the Zeeman effect competes with the Kondo effect, and

$$\delta_\pm \sim \pi/2 \pm \Delta\delta \tag{10}$$

with a certain phase $\Delta\delta$. This phase shift is peculiar to the Zeeman splitting of the Kondo state; the center of the splitting peaks is fixed at the Fermi level in the leads. Pumped charge Q_σ depends on the spin σ accordingly.

Now we investigate the pumping in the Kondo regime along the path shown in Fig. 1(c), where Q_σ is given by

$$Q_\sigma = \frac{e}{2\pi}[2(\delta_{2,\sigma} - \delta_{1,\sigma}) - (\sin 2\delta_{2,\sigma} - \sin 2\delta_{1,\sigma})] \tag{11}$$

with $\delta_{j,\sigma} = \delta_\sigma(V_j)(j = 1, 2)$. In Fig. 3(b), the pumped charge $Q_c = Q_+ + Q_-$ (the dashed line) and pumped spin $Q_s = Q_+ - Q_-$ (the solid line) are plotted as a function of V_1 for a fixed $V_2 = -2\Gamma$ ($-2\Gamma < V_1 < -\Gamma$). We obtain $Q_c \sim 0$ and $Q_s \neq 0$; the spin pumping without the charge pumping [2,12–18] is realized. This result is explained by Eqs. (10) and (11); The effect of $\Delta\delta$ is cancelled for Q_c and doubled for Q_s.

The absence of charge pumping is a generic feature in the Kondo regime, and in contrast to the conductance, which is the maximum of $2e^2/h$. On the other hand, the finite pumped spin proves that the spin degree of freedom is active. These results are consistent with the fact that in the Kondo regime, the charge excitations freeze while the spin excitations are active. Thereby the pumping seizes this separation peculiar to the Kondo effect.

We further demonstrate this separation in the pumping, investigating the pumping along the path shown in Fig. 4(a); tunneling barriers are oscillating alternatively. This pumping path produces zero direct current but finite alternating current. The amplitude of this alternating

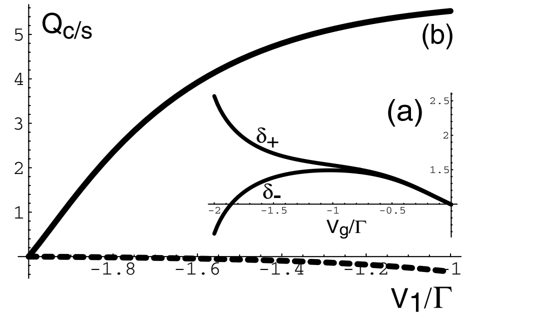

FIG. 3. Electron pumping under the Zeeman effect; $E_Z = 0.5 \times 10^{-3}\Gamma$. (a) δ_σ as a function of the gate voltage V_g. (b) The pumped charge (spin) $Q_{c/s} = Q_+ \pm Q_-$ [dashed (solid) line], (in the unit of $e/2\pi$) for the path in Fig. 1(c) as a function of V_1 for $V_2 = -2.0\Gamma$.

current is proportional to $A_\sigma \equiv 2\sin(2\delta_\sigma)$ since $\frac{d\delta_\sigma}{dt} = 0$. (See Eq. (8).) In Fig. 4(b), the amplitudes of the charge and spin current, $A_c = A_+ + A_-$ (the dashed line) and $A_s = A_+ - A_-$ (the solid line), are plotted as a function of V_1. Around $V_1 \sim 0$, A_c has a peak and decreases to zero as V_1 decreases. On the other hand, A_s is zero for high gate voltage and increases as V_1 decreases, and it has a peak around $V_1 \sim -2\Gamma$.

The separation of the peak structures between A_c and A_s is a direct evidence of the separation of the spin and charge excitations peculiar to the Kondo effect. The peak of A_s appears in the Kondo regime described above. On the other hand, the peak of A_c appears in the mixed-valence regime, where the number of electrons in the dot can fluctuate; the charge excitations are active. In this way, the pumping reveals the intrinsic nature of electronic states in the Kondo effect. Note that when $V_1 \ll -\Gamma$, where the spin-polarized state appears, both $A_{s/c}$ are zero.

Keldysh Green functions.—We now discuss the derivation of Eqs. (4) and (5). Equation (3) is expressed in terms of the Keldysh Green functions [36]: $I_\sigma = I_{\sigma,B} + I_{\sigma,F}$ with $I_{\sigma,B} = \frac{2ieV\xi\eta}{4\hbar} V(G^R_\sigma g^<_{\sigma A} + g^<_{\sigma A} G^A_\sigma)$

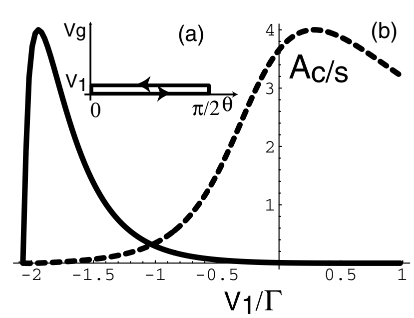

FIG. 4. (a) A pumping path in the (θ, V_g) plane. (b) The amplitude of the charge (spin) alternating current, $A_{c/s} = A_+ \pm A_-$ [dashed (solid) line] as a function of V_1.

and, $I_{\sigma,F} = \frac{eV(\xi^2-\eta^2)}{4\hbar} \times V[G^R_\sigma g^<_{\sigma S} - g^<_{\sigma S} G^A_\sigma + G^<_\sigma g^R_{\sigma S} - g^A_{\sigma S} G^<_\sigma]$, where we have introduced the Keldysh Green functions in the dot and leads, G^j_σ and $g^j_{\sigma k s}$ ($j = R, A, <$), and $g^j_{\sigma S/A} = \sum_k (g^j_{\sigma k+} \pm g^j_{\sigma k-})$. The notation of $VG^R_\sigma g^<_{\sigma A}$ stands for $\int dt_1 V(t_1) G^R_\sigma(t, t_1) g^<_{\sigma A}(t_1, t)$ for example.

To investigate adiabatic pumping regime, we have assumed the following four points: (a) $g^<_{\sigma k s}(t,t') = g^<_{\sigma k s}(t-t';t) = if[\epsilon_k - \mu(t) - s\hbar a(t)]\exp[-i\epsilon_k/\hbar(t-t')]$, which means that the leads keep thermal equilibrium with the time-dependent chemical potential $\mu(t) \pm \hbar a(t)$. (b) $G^{R/A}_\sigma(t,t') = G^{R/A}_\sigma(t-t';t) = \mp i\theta(\pm t \mp t') \times \exp\{-[iE_\sigma \pm \Gamma(t)]/\hbar(t-t')\}$, the Fourier transformation of which is given just below Eq. (5). (c) $V(t)V(t') \simeq V(t)^2$ under the integration. (d) $f[\epsilon - \mu(t) - s\hbar a(t)] \simeq f[\epsilon - \mu(t)] - \frac{\partial f[\epsilon - \mu(t)]}{\partial \epsilon} s\hbar a(t)$. The first three assumptions rely on the relation of $\Omega \ll \Gamma(t)/\hbar$ and the system follows instantaneous values of $V_{L/R}$ and μ.

The above assumptions simplify the expressions of $I_{\sigma,B}$ as in Eq. (4) [37]. By the assumption (d), $I_{\sigma,F}$ is independent of $a(t)$ and accordingly the resulting expression represents the current between the dot and double leads that have a common chemical potential $\mu(t)$. Then we can disregard the current between the two leads in the expression, and it is always detected by the change of $n_{\sigma;\mathrm{dot}}$. In general, the first two terms in $I_{\sigma,F}$ represent the current from the leads to the dot while the rest two terms represent the current from the dot to leads [36]. The sum of two terms therefore yields the time derivative of $n_{\sigma;\mathrm{dot}}$. Note that we can finally apply the Friedel sum rule [32] to relate $n_{\sigma;\mathrm{dot}}$ with δ_σ at this point.

In conclusion, we have investigated adiabatic electron pumping with time-dependent barriers under the electron interactions and shown the separation of the pumped charge and spin peculiar to the Kondo effect. The adiabatic pumping will be a new probe to disclose the electron interactions, which is not achieved by conventional conductance measurements.

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