Edge Singularity for the Optically Induced Kondo Effect in a Quantum Dot

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We analyze the optically induced Kondo effect in the absorption spectrum for a quantum dot with an even number of electrons, for which the Kondo effect does not occur in the ground state. The Kondo exchange couplings generated for photo-excited states can be either antiferromagnetic or ferromagnetic, which may result in different types of the edge singularities, depending on the microscopic parameters of the dot. We discuss critical properties in the spectrum by using low-energy effective field theory.

Recently quantum dots have attracted much attention. They provide useful systems to study the effects of local electron correlations, for which the microscopic parameters can be varied systematically. For instance, the Kondo effect found in resonant tunneling phenomena has been intensively studied theoretically [1–5] and experimentally [6–8]. Besides such transport experiments, optical probes are also useful to explore many-body effects in quantum dots. Although optical experiments have revealed a number of interesting properties, they have been mainly concerned with the properties of charge excitations. Magnetic correlations in the optical spectrum for quantum dots have not been studied systematically thus far. Recently, Kikoin et al. discussed how the spin degrees of freedom can affect the optical line shape for a quantum dot with an even number of electrons, for which the Kondo-type correlation is developed in photo-excited states [8]. In a similar context, it was demonstrated that such Kondo-type correlations may appear in photoemission experiments, resulting in anomalous edge-singularity properties in the spectrum [8]. It was also pointed out that the non linear optical response is affected by these spin correlations [10]. These Kondo-type effects in dynamical quantities such as the optical conductivity and the photoemission spectrum, may be referred to as the dynamically induced Kondo effect.

In this paper we study the dynamically induced Kondo effect in the optical absorption spectrum of a quantum dot having two levels with no spin moment in the ground state. We show that, in the optically excited state where each level is occupied by one electron, the effective spin moment in each level has a Kondo exchange coupling to the leads, which can be either antiferromagnetic or ferromagnetic. We use an effective Kondo model to analyze low-energy critical properties in the optical absorption spectrum around the particle-hole excitation energy of the dot. We find that the effective Kondo coupling leads to strongly field-dependent edge singularity.

We start with a two-level quantum dot which has two leads attached. This system is described by the impurity Anderson model involving two orbitals with the Hamiltonian

\[
H = \sum_{b\alpha} \varepsilon_{b\alpha} c_b^\dagger c_b + \sum_{\alpha} \varepsilon_{\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow} + U \sum_{\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow} + U' \sum_{\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow} + \sum_{b b' \alpha \sigma} (V_{b\alpha} c_b^\dagger d_{b'\alpha} + \text{h.c.}),
\]

where \(c_{b\alpha} (c_{b\alpha}^\dagger)\) annihilates an electron in the left (right) leads, \(d_{b\alpha}\) annihilates an electron in the orbital \(\alpha\) in the dot \((\alpha = 1, 2)\), and \(n_{\alpha\sigma} = c_{b\alpha}^\dagger c_{b\alpha} d_{b\alpha}^\dagger d_{b\alpha}\). Both intra- and inter-orbital Coulomb repulsion in the dot are included. The last term with the coupling \(V_{b\alpha}\) describes the tunneling between the dot and the leads and will be denoted as \(H_T (H = H_0 + H_T)\). We will consider the optical transition from the state where only the lower \((\alpha = 1)\) orbital is occupied by two electrons to a state where each orbital is occupied by one electron. The spin of each orbital in the excited state is subject to the Kondo exchange interaction with the electrons in the leads. The antiferromagnetic coupling of our main interest, the presence of two leads allows the screening of both spins by two orthogonal channels of the lead electrons. The effective decoupling of screening channels becomes immediately clear for the case where the two Kondo states have very different Kondo temperatures and one spin is completely screened at a much higher energy than the other. The residual Kondo coupling leads then to the Kondo effect for the second spin. Therefore, without losing generality we may use a simple illustrative model where the two channels coupled to the two spins are independent and orthogonal to each other in an obvious form already in the bare Hamiltonian. We assume that the quantum dot including leads has the reflection symmetry with respect to a mirror plane in the center, so that we may classify states by parity. Note that this is also compatible with the requirement of an optical (dipole) transition. We thus assume that the hybridizations \(V_{b\alpha}\) is either symmetric or antisymmetric: \(V_{b\uparrow} = V_{b\downarrow} \equiv V_1/(2\sqrt{2})\), \(V_{b\downarrow} = -V_{b\uparrow} \equiv V_2/(2\sqrt{2})\).

We are interested in the ground-state configuration where two electrons occupy the lower level \((\alpha = 1)\) in the dot. This is realized as a lowest energy state when the conditions

\[
\varepsilon_2 > \varepsilon_1 + U - U', \quad \varepsilon_1 < -U, \quad \varepsilon_2 > -2U'
\]
The optical transition is caused by the operator $O$, $|\alpha\rangle$ is the excited state and $\Gamma_0$ is the level width with $D$ being the band width in the leads. Since there is no effective spin moment in the dot, the Kondo effect does not occur in the ground state. As demonstrated by Kikoin et al. [8,9], however, the Kondo effect shows up in optical absorption spectra, whose critical behavior will be studied in detail below.

The optical absorption spectrum of the dot is given by

$$I(\omega) = -2|M|^2 \text{Im} G^>(\omega + i\delta),$$  \hspace{1cm} (3)$$

where $\delta$ is positive infinitesimal and $M$ represents the optical transition matrix element between the two quantum dot states. The dynamical correlation function, $G^>$, is expressed as

$$G^>(\omega) = \frac{1}{\omega - \Delta_0} + \frac{1}{(\omega - \Delta_0)^2} \langle 0 | OT(\omega) O^{|0}. \hspace{1cm} (4)$$

With the conditions (2), the ground state $|0\rangle$ is given by $|0\rangle = d^+_1 d^+_1 [F]$, where $|F\rangle$ is the filled Fermi sea.

The optical transition is caused by the operator $O^I = (1/\sqrt{2}) \sum_{\alpha} \epsilon_{L\sigma} a^\dagger_{\alpha\sigma} a_{\alpha\sigma}$. Up to second order in $H_T$ the $T$-matrix, defined by $T(\omega) = H_T + H_T(\omega - H_0)^{-1} H_T + \cdots$, is reduced to the effective Hamiltonian for the excited state $O^{|0},$

$$H_{\text{eff}} = \sum_{\alpha} \left( \frac{\Gamma_0}{\pi} \ln \frac{D}{-\epsilon_{\alpha} - U} + V_{\alpha\sigma} \sum_{\sigma'} \psi^\dagger_{\alpha\sigma}(0) \psi_{\alpha\sigma'}(0) + 4\pi J_{K}\sum_{\alpha\sigma\sigma'} \psi^\dagger_{\alpha\sigma}(0) \frac{\sigma_{\alpha'}^\dagger}{2} \psi_{\alpha\sigma'}(0) S^\dagger_{\alpha} \right).$$

Here $\sigma^\dagger$ is the Pauli matrix, and $S^\dagger_{\alpha} = (1/2) d^\dagger_{\alpha} \sigma^\dagger_{\alpha\sigma} d_{\alpha\sigma}$ are effective impurity spins formed in the two levels in the excited state $O^{|0}$. The impurity spins are coupled to electrons in the leads, $\psi_{1\sigma}(0) = (1/\sqrt{2}) \sum_k (c^\dagger_{k\sigma} + c_{k\sigma})$, $\psi_{2\sigma}(0) = (1/\sqrt{2}) \sum_k (c^\dagger_{k\sigma} - c_{k\sigma})$, via two kinds of Kondo exchange interactions

$$J_{K\alpha} = |V_{\alpha}|^2 \left( \frac{1}{\epsilon_{\alpha} + U'} - \frac{1}{\epsilon_{\alpha} + U'} + \frac{1}{\epsilon_{\alpha} + U'} \right). \hspace{1cm} (5)$$

Note that the Kondo couplings can be either antiferromagnetic or ferromagnetic because the energy of intermediate states measured from the optically excited states, $-\epsilon_{\alpha} - U'$ and $\epsilon_{\alpha} + U'$, are not necessarily positive. The Kondo couplings cause final-state interactions as in the Fermi-edge singularity [4,13], giving rise to interesting features in the absorption spectrum as we will discuss below. The parameter region of our interest is shown in Fig. 1. We note that an antiferromagnetic Kondo coupling is not generated without the inter-orbital Coulomb interaction $U'$, in which case the Kondo screening does not occur in the optically excited state.

We will analyze the critical behavior in the optical absorption spectrum in detail for the following two cases: (i) $J_{K1} > 0$ and $J_{K2} > 0$ and (ii) $J_{K1} < 0$ and $J_{K2} > 0$. We will first ignore the potential scattering term $V_{\alpha\sigma}$ and concentrate on the dynamical properties of spin degree of freedom. The potential scattering affects only the charge sector and its effect will be discussed shortly. The Kondo effect in $H_{\text{eff}}$ is none but two decoupled single-channel Kondo problems, whose low-energy physics can be studied in the standard manner using the one-dimensional (1D) model for electrons in the leads [16,17]. We thus introduce two left-going electron modes with linear dispersion $\psi_{\alpha L\sigma}(x)$ defined on a full line $(-\infty < x < \infty)$, which satisfy $\psi_{\alpha L\sigma}(0) = \psi_{\alpha\sigma}(0)$. We then use nonabelian bosonization methods to rewrite the Hamiltonian. In this representation the charge and spin sectors are decoupled and the spin part of the kinetic energy of 1D electrons reads

$$\mathcal{H}_0 = \frac{v_F}{2\pi} \sum_{\alpha} \int \frac{dx}{k + 2} \frac{1}{k^2 + 2} : J^a_{\alpha L}(x) J^a_{\alpha L}(x) :,$$ \hspace{1cm} (6)$$

where $v_F$ is the Fermi velocity and the level $k = 1$ SU(2) currents are defined by

$$J^a_{\alpha L}(x) = \sum_{\sigma\sigma'} : \psi^\dagger_{\alpha L\sigma}(x) \sigma^a_{\sigma\sigma'} \psi_{\alpha L\sigma}(x) :.$$

The Kondo interactions in $H_{\text{eff}}$ is written

$$\mathcal{H}_K = \sum_{\alpha} \lambda_{K\alpha} J^a_{\alpha L}(0) S^a_{\alpha\sigma},$$ \hspace{1cm} (8)$$

where $\lambda_{K\alpha} = 4\pi J_{K\alpha} / v_F$. Finally the optical absorption spectrum $I(\omega)$ is given by the Fourier transform of

$$G(t) = \langle 0 | O e^{\mathcal{H}_0 t} e^{-i(\mathcal{H}_0 + \mathcal{H}_K) t} O^{|0} e^{-i\Delta t},$$ \hspace{1cm} (9)$$

where

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1}
\caption{Energy-level diagram specified by the two Kondo exchange couplings $J_{K\alpha}$ which can be either positive or negative within the conditions [4]. For example, in the region denoted as AF, $J_{K1}$ is antiferromagnetic and $J_{K2}$ is ferromagnetic.}
\end{figure}
\[ \Delta = \Delta_0 - \sum_{\alpha} \frac{\Gamma_{\alpha}}{\pi} \ln \frac{D}{-\varepsilon_{\alpha} - U} \] (10)

is the renormalized particle-hole excitation energy.

(i) region \( AA \) with \( J_{K1} > 0 \) and \( J_{K2} > 0 \). The weak antiferromagnetic Kondo couplings are renormalized to the strong-coupling fixed point, where \( \lambda_{K\alpha} \to \lambda^*_{K\alpha} = 2/(k + 2) \) with \( k = 1 \). Following Refs. [15, 17], we introduce the unitary operators \( U_{\alpha} \) transforming the \( SU(2) \) currents as \( U_{\alpha} J_{\alpha L}^a(x) U_{\alpha}^\dagger = J_{\alpha L}^a(x) + 2\pi S_0^z \delta(x) \equiv J_{\alpha L}^a(x) \). The unitary operators \( U_1 \) and \( U_2 \) commute with each other because the Hamiltonian is decoupled into two sectors. The merit of using \( U \) is that it allows us to rewrite the Hamiltonian as

\[ U\mathcal{H}_0 U^\dagger = \mathcal{H}_0 + \mathcal{H}_K \] (11)

at \( \lambda_{K\alpha} = \lambda^*_{K\alpha} \). The long-time behavior of \( G(t) \) is then obtained from the correlation function of \( U_{\alpha} \),

\[ G(t) \sim \langle U_1(t) U_{\alpha L}^0(0) \rangle \langle U_2(t) U_{\alpha R}^0(0) \rangle e^{-\Delta t} \] (12)

where \( U_{\alpha}(t) = e^{i\mathcal{H}_0 t} U_{\alpha} e^{-i\mathcal{H}_0 t} \) can be regarded as a boundary condition changing operator [13, 13] and the brackets represents the average at the strong-coupling fixed point. We may rewrite (11) by using the operator product expansion (OPE) in terms of the new currents \( \mathcal{J}_{\alpha L}^a \):

\[ \mathcal{J}_{\alpha L}^a(z) U_{\alpha L}^0(w) = \frac{-S_0^a}{z - w} U_{\alpha L}^0(w) + \text{reg.} \cdots, \] (13)

which just corresponds to the OPE for the highest weight of \( SU(2) \) current algebra. Accordingly, the correlation function for \( U_{\alpha L}^0(w) \) satisfies the so-called Knizhnik-Zamolodchikov equation [14],

\[ \left( \frac{\partial}{\partial z_i} + \frac{2}{k + 2} \sum_{\alpha \neq i} \frac{S_{\alpha}^a \otimes \bar{S}_{\alpha}^a}{z_i - z_l} \right) \langle U_{\alpha} (z_i) U_{\alpha L}^0 (z_l) \rangle = 0, \] (14)

where the operator \( \bar{S}_{\alpha}^a \) associated with \( U_{\alpha} \) acts from the right like \( S_{\alpha}^a \cdot U_{\alpha}(z_i) = U_{\alpha}(z_i) \bar{S}_{\alpha}^a \) whereas \( S_{\alpha}^a \) associated with \( U_{\alpha L}^0 \) acts from the left. Since the impurity spin \( S_0^a \) satisfies \( \sum_{\alpha} S_{\alpha}^a S_{\alpha}^a = S(S + 1) \), the solution of Eq. (14) is

\[ \langle U_{\alpha} (t) U_{\alpha L}^0 (0) \rangle = \frac{1}{t^{2\Delta_s}} \] (15)

with the boundary dimension,

\[ \Delta_s = \frac{S(S + 1)}{k + 2}. \] (16)

For the present model \( (S = 1/2, k = 1) \), \( \Delta_s = 1/4 \) \([14] \) for each channel \( \alpha \).

The Kondo temperature can be different for two channels \([12] \), depending on the parameters \( \varepsilon_{\alpha} \), \( U \), and \( \Gamma_{\alpha} \).

In general we can expect two critical regions to appear in the absorption spectrum:

\[ I(\omega) = \left\{ \begin{array}{ll} (\omega - \Delta)^{4\Delta_s - 1}, & 0 < \omega - \Delta < T_{K1}, \\ (\omega - \Delta)^{2\Delta_s - 1}, & T_{K1} < \omega - \Delta < T_{K2}. \end{array} \right. \] (17)

For typical values of small quantum dots we may encounter a situation where, say, \( T_{K1} < T_{K2} \) because the deeper level may have a smaller \( \Gamma_1 \). In this case the impurity spin \( S_1 \) remains essentially free whereas the spin \( S_2 \) is screened, and the Kondo effect with the critical behavior \( (\omega - \Delta)^{-1/2} \) may be observed in the low-energy regime of the absorption spectrum. A twist to the argument occurs when we introduce Hund’s-rule coupling \( J_H S_1^z S_2^z \) between the spins of the two orbitals \([21] \). In this case the coupling will generate a logarithmic correction to the \( (\omega - \Delta)^{-1/2} \) behavior in \( T_{K1} \approx T \approx T_{K2} \), as we will show later \([\text{Eq. (21)}] \).

So far we have exploited \( SU(2) \) current algebra to discuss low-energy properties of the absorption spectrum. Although this analysis cannot be used when the magnetic field is applied, we can still evaluate the boundary dimension \( \Delta_s \) as a function of magnetic fields, by applying the finite-size scaling to the exact solution of the Kondo model \([21, 22] \). Namely, the scaling dimension \( \Delta_s \) is determined by analyzing the excitation spectrum in the presence of the Kondo impurity. Following the analysis in \[3] \), we obtain

\[ \Delta_s = n_{\text{imp}}^2, \quad n_{\text{imp}} = \int_{-\infty}^{\Lambda_0} \sigma_{\text{imp}}(\Lambda) d\Lambda. \] (18)

Here \( \Lambda_0 = -1/J + (1/\pi) \ln(\sqrt{2}T_H/H) \) and \( \sigma_{\text{imp}} \) is determined by the integral equation for the exact solution of the Kondo model \([21, 22] \),

\[ \sigma_{\text{imp}}(\Lambda) = \frac{1}{2\pi[(\Lambda + 1/J)^2 + 1/4]} - \int_{-\infty}^{\Lambda_0} \sigma_{\text{imp}}(\Lambda') d\Lambda'. \]

The critical exponent thus obtained for the absorption spectrum is shown in Fig. 2 as a function of the magnetic field.
At $H = 0$, we recover $\Delta_s = 1/4$, obtained above by current algebra techniques. When the potential scattering terms $V_{\alpha\beta}$ in $H_{\text{eff}}$ neglected so far is incorporated into $G(t)$, where the scaling dimension $\Delta_c$ for the charge sector is determined by the phase shift due to the potential scattering. The divergence singularity in the spectrum is weakened by the additional exponent $2\Delta_c$. Since $\Delta_c$ has little dependence on the magnetic field, the field-dependence of the critical exponent is solely determined by the dynamically induced Kondo effect, as shown in Fig. 2.

(ii) region $FA$ with $J_{K1} < 0$ and $J_{K2} > 0$. We now turn to the case where only one of the Kondo couplings is antiferromagnetic. In this case, the impurity spin $S_1$ is decoupled from the lead electrons at the fixed point because the ferromagnetic Kondo coupling is marginally irrelevant, whereas the impurity spin $S_2$ is completely screened due to the Kondo effect. Note that a somewhat similar situation occurs in the intermediate temperature range $T_{K1} \ll T \ll T_{K2}$ for the case discussed above, where one spin is screened while the other is still basically free. With the inclusion of the Hund’s-rule coupling between $S_1$ and $S_2$, the two spins may be effectively combined to form a spin triplet $S = 1$, which is then screened by only one channel ($\alpha = 2$) of electrons, the so-called underscreened Kondo system is realized. It is known that for the fixed point of this system the decoupled impurity $S_1$ weakly interacts with the renormalized current $J_{2L}$ via the ferromagnetic Kondo interaction, $\lambda_3 J_{2L}^\alpha (t) S_1^\alpha$. Although the coupling is marginally irrelevant and leads to $\langle U_1(t) U_1^\dagger (0) \rangle \sim \text{const.}$, it still affects the long-time behavior of $\langle U_2(t) U_2^\dagger (0) \rangle$. We can explicitly evaluate it around the fixed point, e.g., by solving Callan-Symanzik equation in the renormalization group,

$$\left( \frac{\partial}{\partial \ln t} + 2\gamma_2 - \beta_2 \frac{\partial}{\partial \alpha_2} - \beta_3 \frac{\partial}{\partial \alpha_3} \right) \langle U_2(t) U_2^\dagger (0) \rangle = 0.$$  

Here $\beta_2$ is the beta function for $\lambda_2 U_2$ whereas $\beta_3$ is that for the effective ferromagnetic coupling $\lambda_3 J_{2L}^\alpha (t) S_1^\alpha$ caused by the Hund’s-rule coupling. The corresponding scaling equations read

$$\beta_2 = \frac{d \beta_2}{d \ln t} = 3 \lambda_2 - \frac{1}{4} \lambda_2 \lambda_3,$$

$$\beta_3 = \frac{d \beta_3}{d \ln t} = \lambda_3^2 - \frac{1}{4} \lambda_2^2,$$

where $\gamma_2 \equiv 1 - \frac{\partial \ln \beta_2}{\partial \ln \alpha_2}$. By solving these equations, we have the leading-order contribution

$$\langle U_2(t) U_2^\dagger (0) \rangle \propto \sqrt{\ln \frac{1}{t}}.$$  

This leads to the low-energy behavior of the absorption spectra,

$$I(\omega) \propto \sqrt{\frac{1}{\omega - \Delta}} \ln \left( \frac{1}{\omega - \Delta} \right) \text{ for } 0 < \omega - \Delta < T_{K2}. \tag{21}$$

The logarithmic correction appears due to the marginally irrelevant ferromagnetic coupling.

In summary, we have studied the edge singularity in the optically induced Kondo effect for a quantum dot with an even number of electrons. We have shown that the Kondo exchange couplings generated in photo-excited states can be either antiferromagnetic or ferromagnetic, which may lead to different anomalous behaviors in the optical absorption spectrum according to the microscopic parameters for a quantum dot. It is interesting to observe such edge singularity in the absorption spectrum experimentally by properly tuning the physical parameters such as the gate voltage and the magnetic field.

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