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Kondo Effect in Superconducting Nanoparticles

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We investigate the Kondo effect and superconductivity in ultrasmall grains by using a model, which consists of *sd* and BCS Hamiltonians with the introduction of a pseudofermion. We discuss physical properties of the condensation energy and behavior of the gap function and the spin singlet order parameter corresponding to the Kondo effect in relation to the critical level spacing and co-existence. We find that strong local magnetic moments from the impurities makes the transition temperature for superconductivity reduced. However, weak couplings λ of the superconductivity do not destroy the spin singlet order parameter at all. Finally we derive the exact equation for the Kondo regime in nanosystem and discuss the condensation energy from the viewpoint of the energy level.

Keywords:

1. INTRODUCTION

The Kondo effect has been much attracted great interest in the properties in semiconductor quantum dots. The Kondo effect can be understood as a magnetic exchange interaction between a localized impurity spin and free conduction electrons.[1] To minimize the exchange energy, the conduction electrons tend to screen the spin of the magnetic impurity and the ensemble forms a spin singlet. In a quantum dot, some exotic properties of the Kondo effect have been observed.[2–4] Recently, Sasaki et al. has found a large Kondo effect in a quantum dots with an even number of electrons.[5] The spacing of discrete levels in such quantum dots is comparable with the strength of electron–electron Coulomb interaction. The Kondo effect in multilevel quantum dots has been investigated theoretically by several groups.[6–8] They have shown that the contribution from multilevels enhances the Kondo effect in normal metals. There are some investigations on the Kondo effect in quantum dots coupled ferromagnetism,[9] noncollinear magnetism,[10] superconductivity[11] and so on.[12,13]

Properties of ultrasmall superconducting grains have been also theoretically investigated by many groups.[15–24] Black et al. have revealed the presence of a parity dependent spectroscopic gap in tunnelling spectra of nanosize Al grains.[15,16] In such ultrasmall superconducting grains, the bulk gap has been discussed in relation to physical properties such as the parity gap,[21] condensation energy,[22] electron correlation[23] with the dependence of level spacing of samples.[24] In previous works,[25] we have also discussed physical properties such as condensation energy, parity gap, and electron correlation of two-gap superconductivity in relation to the size dependence and effective pair scattering process. The possibility of new two-gap superconductivity has been also discussed by many groups.[26–37]

In a standard s-wave superconductor, the electrons form pairs with antialigned spins and are in a singlet state as well. When the superconductivity and Kondo effect present simultaneously, the Kondo effect and superconductivity are usually expected to be competing physical phenomena. The local magnetic moments from the impurities tend to align the spins of the electron pairs in the superconductor which often results in a strongly reduced transition temperature. Buitelaar et al. have experimentally investigated the Kondo effect in a carbon nanotube quantum dot coupled to superconducting Au/Al leads.[11] The have found that the superconductivity of the leads does not destroy the Kondo correlations on the quantum dot when the Kondo temperature. A more subtle interplay has been proposed for exotic and less well-understood materials such as heavy-fermion superconductors in which both effects might actually coexist.[38]

In this paper, we investigate the Kondo effect and superconductivity in ultrasmall grains by using a model, which consists of sd and reduced BCS Hamiltonians with the introduction of a pseudofermion. A mean field approximation for the model is introduced, and we calculate physical properties of the critical level spacing and the condensation energy. These physical properties are discussed in relation to the coexistence of both the superconductivity and the Kondo regime. Finally we derive the exact equation for the Kondo regime in nanosystem and discuss the condensation energy from the viewpoint of the correlation energy.

2. KONDO REGIME COUPLED TO SUPERCONDUCTIVITY

In nanosize superconducting grains, the quantum level spacing approaches the superconducting gap. It is necessary to treat discretized energy levels of the small system. For ultrasmall superconducting grains, we can consider the pairing-force Hamiltonian to describe electronic structure of the system[39] and can know the critical level spacing where the superconducting gap function vanishes at a quantum level spacing.[24] In this section, we present a model for a system in Kondo regime coupled to superconductivity and discuss physical properties such as critical level spacing and condensation energy by using a mean field approximation in relation to gap function, spin singlet order as the Kondo effect, coexisitence and so on.

2.1. MODEL

We consider a model coupled to superconductivity for quantum dots to investigate the Kondo effect in normal metals, which can be expressed by the effective low-energy Hamiltonian obtained by the Schrieffer-Wolff transformation:[40]

$$H = H_0 + H_1 + H_2 \tag{1}$$

where

$$H_0 = \sum_{k,\sigma} \varepsilon_k a^\dagger_{k\sigma} a_{k\sigma} + \sum_\sigma E_\sigma d^\dagger_\sigma d_\sigma \tag{2}$$

$$H_1 = J \sum_{k,k'} [S_+ a^\dagger_{k'\downarrow} a_{k\uparrow} + S_- a^\dagger_{k'\uparrow} a_{k\downarrow} + S_z (a^\dagger_{k'\uparrow} a_{k\uparrow} - a^\dagger_{k'\downarrow} a_{k\downarrow})] \tag{3}$$

$$H_2 = -g \sum_{k,k'} a^\dagger_{k\uparrow} a^\dagger_{k\downarrow} a_{k'\downarrow} a_{k'\uparrow} \tag{4}$$

$a^\dagger_{k\sigma}$ ($a_{k\sigma}$) and $d^\dagger_\sigma$ (d_σ) are the creation (annihilation) operator corresponding to conduction electrons and the effective magnetic particle as an impurity, respectively. In this study we assume the magnetic particle is fermion $S = 1/2$ for the simplicity. E means an extraction energy given by $E_{\uparrow,\downarrow} = -E_0 \pm E_z$ included the Zeeman effect. The second term in Eq. (1) means the interaction between conduction electrons and the spin in a quantum dot. S is the spin operator as $S_+ = d^\dagger_\uparrow d_\downarrow$, $S_- = d^\dagger_\downarrow d_\uparrow$, and $S_z = (d^\dagger_\uparrow d_\uparrow - d^\dagger_\downarrow d_\downarrow)/2$. The third term corresponds to the interaction between conduction electrons from the pairing force Hamiltonian.

Here, we introduce a pseudofermion for the magnetic particle operator[41] as

$$d^\dagger_\uparrow = f_\downarrow, \quad d_\uparrow = f^\dagger_\downarrow, \quad d^\dagger_\downarrow = -f_\uparrow, \quad d_\downarrow = -f^\dagger_\uparrow \tag{5}$$

In this transformation, we have the condition

$$f^\dagger_\uparrow f_\uparrow + f^\dagger_\downarrow f_\downarrow = 1 \tag{6}$$

We can know $|\sigma\rangle = f^\dagger_\sigma |0\rangle$. The spin operator S can be rewritten as $S_+ = f^\dagger_\uparrow f_\downarrow$, $S_- = f^\dagger_\downarrow f_\uparrow$, and $S_z = (f^\dagger_\uparrow f_\uparrow - f^\dagger_\downarrow f_\downarrow)/2$. The Hamiltonian can be rewritten as

$$H_0 = \sum_{k,\sigma} \tilde{\varepsilon}_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_\sigma E f^\dagger_\sigma f_\sigma \tag{7}$$

$$H_1 = J \sum_{k,k',\sigma,\sigma'} f^\dagger_\sigma f_{\sigma'} c^\dagger_{k'\sigma'} c_{k\sigma} \tag{8}$$

$$H_2 = -g \sum_{k,k'} c^\dagger_{k\uparrow} c^\dagger_{k\downarrow} c_{k'\downarrow} c_{k'\uparrow} \tag{9}$$

where $c_{k\sigma} = \sum_i U_{ik} a_{i\sigma}$ with $\tilde{\varepsilon}_k = \sum_{i,j} U^\dagger_{ki} [\varepsilon_i \delta_{ij} - J/2] U_{jk}$. For the simplicity, we only focus $E_z = 0$ without an external magnetic field: $E = E_0$.

2.2. Mean Field Approximation

In this section, we introduce a mean field approximation for the present Hamiltonian of Eq. (1). Eto et al. have presented the mean field approximation for the Kondo effect in quantum dots.[42]

In the mean field approximation, we can introduce the spin singlet order parameter

$$\Xi = \frac{1}{\sqrt{2}} \sum_{k,\sigma} \langle f^\dagger_\sigma c_{k\sigma} \rangle \tag{10}$$

This order parameter describes the spin couplings between the dot states and conduction electrons. The superconducting gap function can be expressed as

$$\Delta = \sum_k \langle c_{k\downarrow} c_{k\uparrow} \rangle \tag{11}$$

Using these order parameters in Eqs. (8) and (9), we obtain the mean-field Hamiltonian

$$H_{\mathrm{MF}} = \sum_{k,\sigma} \tilde{\varepsilon}_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_\sigma \tilde{E} f^\dagger_\sigma f_\sigma + \sqrt{2} J \sum_{k,\sigma} [\Xi f_\sigma c^\dagger_{k\sigma} + \Xi^* c_{k\sigma} f^\dagger_\sigma] - g \sum_k [\Delta^* c_{k\downarrow} c_{k\uparrow} + \Delta c^\dagger_{k\uparrow} c^\dagger_{k\downarrow}] \tag{12}$$

The constraint of Eq. (6) is taken into account by the second term with a Lagrange multiplier λ. In this study, we assume a constant density of state with the energy region of the Deby energy, and the coupling constants can be expressed as $J = d\tilde{J}$ and $g = d\lambda$.

3. DISCUSSION

By minimizing the expectation value of H_{MF} in Eq. (12), the order parameters are determined self-consistently. First, we show the Kondo effect without the pairing force part ($g = 0$) in the framework of the mean field approximation. Next, the Kondo effect in the presence of the superconductivity is discussed in relation to the critical level spacing and condensation energy. Finally, we derive the exact equation for the Kondo effect in ultrasmall grains coupled to normal metals and discuss properties such as the condensation energy in relation to Richardson's exact equation for the superconductivity.

3.1. Critical Level Spacing in Kondo Effect

In ultrasmall grains such as quantum dots etc., the quantum level spacing approaches order parameters. For ultrasmall superconducting grains, the critical level spacing d_c^{BCS} can be expressed as $d_c^{\mathrm{BCS}} = 4\omega_D e^\gamma \exp(-1/\lambda)$ for even number of electrons, where ω_D means the Deby energy. This result suggests that the gap function of a nanosize system with the level spacing d vanishes, when the coupling parameter λ_c is less than the value $(\ln 4\omega_D/d + \gamma)^{-1}$. The bulk gap function Δ_c with λ_c can be expressed as $\Delta_c = \omega_D \mathrm{sh}^{-1}(1/\lambda_c)$.

Figure 1(a) shows the gap function of a nanosize system in the framework of the standard BCS theory. We can find the region where the gap function vanishes, when the coupling becomes less than λ_c. This means the level spacing is larger than gap function in this region.

Here we drive the critical level spacing for only the Kondo regime ($\lambda = 0$). The equation determining the singlet order parameter can be expressed as

$$\Xi = \sum_k \frac{\Xi(\xi_k - x)}{(\xi_k - x)^2 + \Xi^2} \tag{13}$$

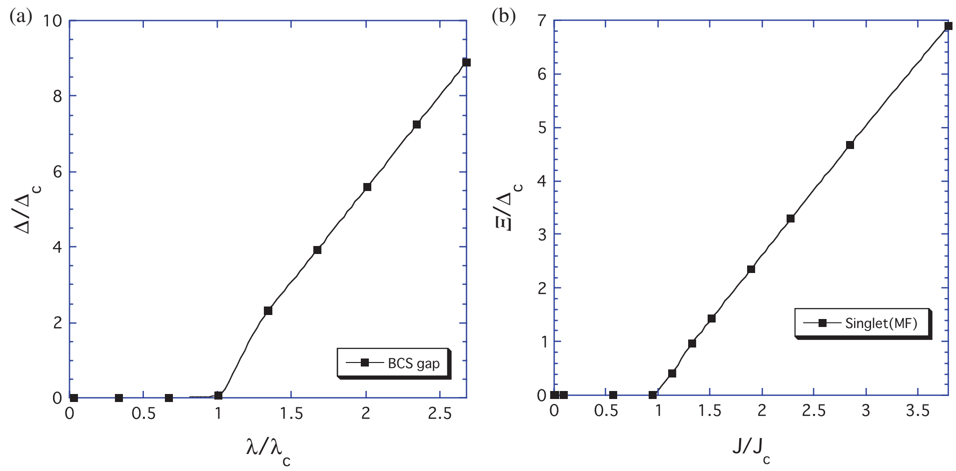

Fig. 1. Gap function and spin singlet order: (a) The gap function. The gap function vanishes in the region of smaller λ value than λ_c. (b) Spin singlet order parameter. In the case of $\tilde{J} < \tilde{J}_c$, the singlet order vanishes. The system consists of 8 energy levels and 8 electrons with the level spacing $d = 1.0$ and $\omega_D = 1.0$.

where $\xi_k = \tilde{\varepsilon}_k - \mu$, $x = [\tilde{\varepsilon}_k + \tilde{E} \pm \sqrt{(\tilde{\varepsilon}_k - \tilde{E})^2 + 4\Xi^2}]]/2$ and μ is the chemical potential. For the case of the critical level spacing, the solution has the spin singlet order vanishes. From Eq. (13), we can find the critical level spacing d_c^{Kondo} for the Kondo regime.

$$d_c^{\text{Kondo}} = 4\omega_D e^{\gamma} \exp\left[-\frac{1}{2\sqrt{2}\tilde{J}}\right] \tag{14}$$

When the coupling parameter $\tilde{J}$ is smaller than $\tilde{J}_c = [2\sqrt{2}(\ln(4\omega_D/d) + \gamma)]^{-1}$, the spin singlet order parameter vanishes.

Figure 1(b) shows the spin singlet order parameter of Eq. (10) in the case $g = 0$. In the region of $\tilde{J} < \tilde{J}_c$, the order parameter vanishes. This result suggests the critical level spacing in the Kondo effect.

3.2. Kondo Effect Coupled to Superconductivity

In this study, we consider a simple system which consists of 8 energy levels and 8 electrons and investigate the critical level spacing and the condensation energy of the coupled system between the superconductivity and the Kondo regime in the framwork of the mean field approximation of Eq. (12).

Figure 2(a) shows the spin singlet order parameter and the gap function for several cases. We can find the critical level spacings for the gap function and for the spin singlet order parameter. When $\lambda < \lambda_c$ and $\tilde{J} > \tilde{J}_c$, we can find only the spin singlet order parameters. In the region of λ/λ_c from 1.4 to 1.7 with $\tilde{J}/\tilde{J}_c = 0.189$, we can find the coexistence of both the gap function and the spin singlet order parameter. In larger λ/λ_c than 1.7, only the gap function still exists, and the spin singlet order parameter vanishes. In $\tilde{J}/\tilde{J}_c = 0.284$, we can find the coexistence in the region $\lambda/\lambda_c = 1.7 - 2.3$. These results suggest that strong local magnetic moments from the impurities makes the transition temperature for superconductivity reduced. However, weak couplings λ of the superconductivity do not destroy the spin singlet order parameter at all. These results are a good agreement with the experimental results.[11] We can find that there is the coexistence region for both the superconductivity and the Kondo regime.

Figure 2(b) shows the condensation energy for several λ and $\tilde{J}$ values. We can find the condensation energy of the coupled

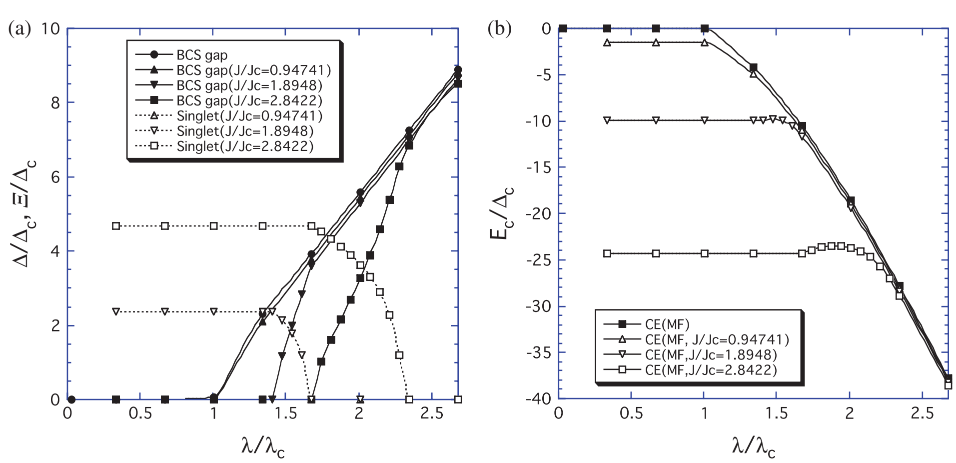

Fig. 2. Physical properties in coupled system: (a) Gap function and spin singlet order parameter. (b) Condensation energy. $\tilde{J}/\tilde{J}_c = 0, 0.94741, 1.8948$, and 2.8422. Other parameters are the same.

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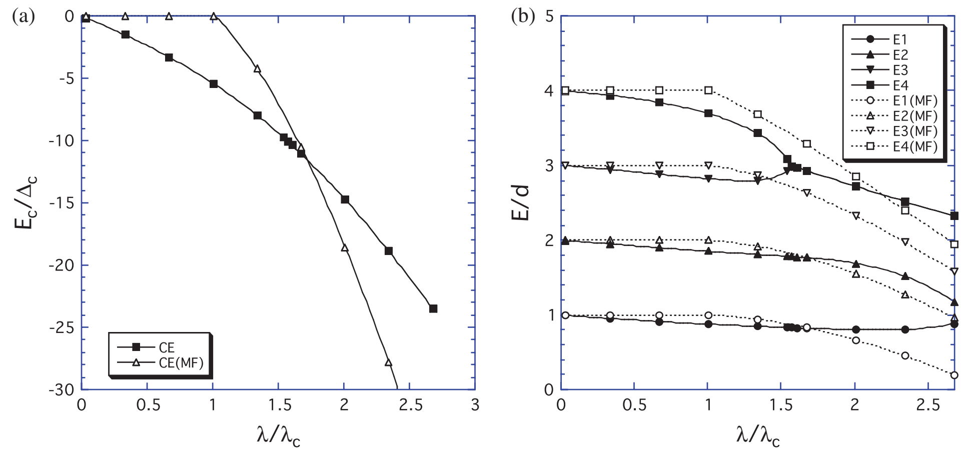

Fig. 3. Exact solution for the superconductivity: (a) Condensation energy of the exact solution with that obtained by the mean field approximation (b) Pairing energy level with energy level obtained by the mean field approximation. 8 energy levels, 8 electrons, $d = 1.0$, $\omega_D = 4.0$.

system between the superconductivity and the Kondo regime becomes lower than that of the pure superconductivity. In the coexistent region, the highest value of the condensation energy appears in all cases.

3.3. Exact Solution for Kondo Regime

The standard BCS theory gives a good description of the phenomenon of superconductivity in large sample. However, when the size of a superconductor becomes small, the BCS theory fails. To investigate physical properties such as the condensation energy, parity gap, etc. it is necessary to take more accurate treatment. For the superconductivity in ultrasmall grains, the exact solution to the reduced BCS Hamiltonian presented by Richardson[39] has been applied to investigate such physical properties.[18]

By using the wave function describing all pair electron excitations, we can derive the exact solution for the pairing force (reduced) Hamiltonian

$$2-\sum_{k=1}^{N}\frac{\lambda}{\tilde{\varepsilon}_k - E_i}+\sum_{l=1,\,l\neq i}^{n}\frac{2\lambda}{E_l - E_i}=0 \tag{15}$$

where N and n are the number of orbital and the number of the occupied orbital, respectively. E_i corresponds to the exact orbital. Figure 3 shows the condensation energy and the pairing energy level for the nanosize superconductivity. Note that physical properties obtained by the mean field approximation give a good description for the high density of state ($d \to \infty$). We can find the different behavior of the condensation energy from that obtained by the mean field approximation as shown in Figure 3(a). Figure 3(b) shows qualitative behaivior of the pairing energy level in the ground state. In λ of about 1.6, above two energy levels in Figure 3(b) are completely paired. The pairing behavior has been already reported by many groups.[39]

Let us derive the exact equation for the Kondo regime in ultrasmall grains. We can consider the Hamiltonian $H = H_0 + H_1$ in Eq. (1). We introduce a creation operator describing all excited states of the spin singlet coupling between a conduction electron and a pseudofermion.

$$B_j^{\dagger}=\sum_{k,\sigma}\frac{c_{k\sigma}^{\dagger}f_{\sigma}}{\tilde{\varepsilon}_k - E_j} \tag{16}$$

where E_j means the exact eigenenergies in the Kondo regime. The exact eigenstate $|\Psi_n\rangle$ for the Kondo regime can be written as $|\Psi_n\rangle = \Pi_{\nu=1}^{n} B_{\nu}^{\dagger}|0\rangle$. Other electrons, which are not related to the spin singlet order, contribute $E_{\text{single}} = \sum_{k=1}^{n}\tilde{\varepsilon}_k$ to the eigenenergy. The ground state energy E_{GS} can be written as $E_{\text{GS}} = \sum_{k=1}^{n}[E_k + \tilde{\varepsilon}_k]$.

By operating the Hamiltonian to the exact eigenstate, we obtain the condition as

$$1+\sum_{k=1}^{N}\frac{\tilde{J}}{\tilde{\varepsilon}_k - E_j}=0 \tag{17}$$

This equation gives the exact solution for the Kondo regime. Note that the creation operator of Eq. (16) might be true boson by comparing with the case of the reduced BCS model.

Figure 4 shows the condensation energy of the exact solution in the Kondo regime with that obtained by the mean field approximation. We can find the different behavior of the condensation energy from that obtained by the mean field approximation. However, the behavior is similar to that in the superconductivity in nanosize system.

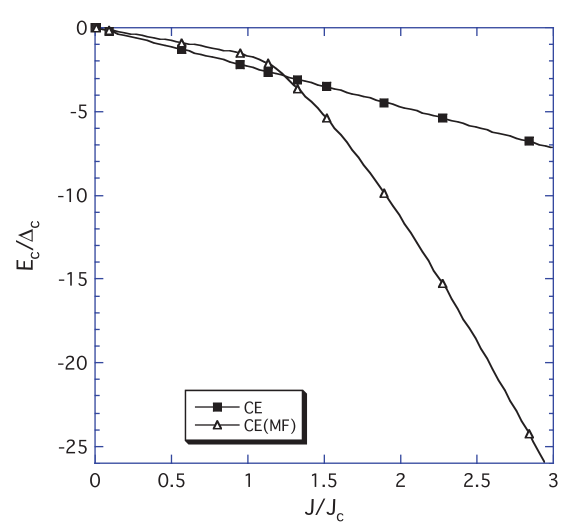

Fig. 4. Condensation energy for the Kondo regime: all parameters used in the system are the same: 8 energy levels, 8 electrons, and $d = 1.0$, $\omega_D = 4.0$.

4. CONCLUDING REMARKS

We have investigated properties of the Kondo regime coupled to the superconductivity in ultrasmall grains by using a mean field approximation. In the framework of the mean field approximation, we have found the critical level spacing for the Kondo regime. The result suggests that the Kondo effect vanishes, when the level spacing becomes larger than the critical level spacing.

We have calculated physical properties of the critical level spacing and the condensation energy of the coupled system by using the mean field approximation. From the results, we have found that strong local magnetic moments from the impurities makes the transition temperature for superconductivity reduced. However, weak couplings λ of the superconductivity do not destroy the spin singlet order parameter at all. These results are a good agreement with the experimental results.[11] We have found that there is the coexistence region for both the superconductivity and the Kondo regime.

Finally we have derived the exact equation for the Kondo regime in nanosystem and have discussed the condensation energy from the viewpoint of the energy level. It might be not easy to find the exact equation for the Kondo regime coupled to superconductivity. The exact properties such as the condensation energy etc. in the Kondo regime by using the exact equation will be presented elsewhere.

In summary, we have investigated the Kondo effect and superconductivity in ultrasmall grains by using a model, which consists of *sd* and reduced BCS Hamiltonians with the introduction of a pseudofermion. A mean field approximation for the model have been introduced, and we have calculated physical properties of the critical level spacing and the condensation energy. These physical properties have been discussed in relation to the coexistence of both the superconductivity and the Kondo regime. Finally we have derived the exact equation for the Kondo regime in nanosystem and discuss the condensation energy from the viewpoint of the energy level.

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