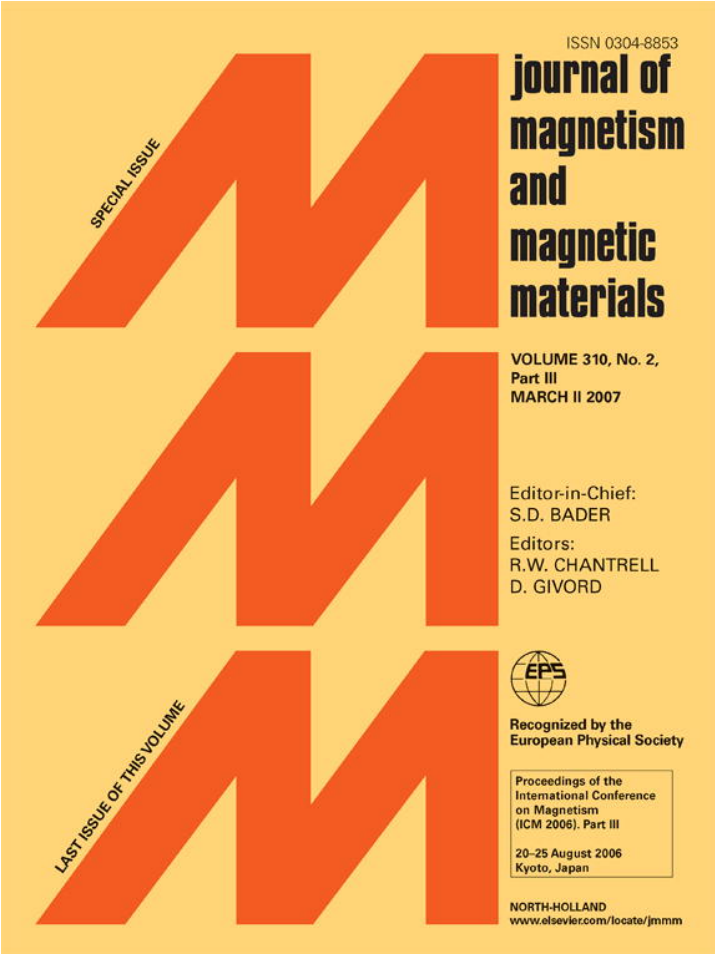
ISSN 0304-8853
journal of magnetism and magnetic materials
SPECIAL ISSUE
VOLUME 310, No. 2, Part III
MARCH II 2007
Editor-in-Chief: S.D. BADER
Editors: R.W. CHANTRELL D. GIVORD
EPS
Recognized by the European Physical Society
Proceedings of the International Conference on Magnetism (ICM 2006). Part III
20–25 August 2006 Kyoto, Japan
LAST ISSUE OF THIS VOLUME
NORTH-HOLLAND
www.elsevier.com/locate/jmmm

Available online at www.sciencedirect.com
ScienceDirect
Journal of Magnetism and Magnetic Materials 310 (2007) 2414–2416
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Kondo effect in organometallic complexes with vibrating ligand shells

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Available online 16 November 2006

Abstract

We investigate transport through a mononuclear rare-earth metal-organic shell complex with strong tunnel coupling between the shell and two electrodes. The ground state of this molecule is a singlet while the first excited state is a triplet. We show that modulation of the tunnel barrier due to a molecular distortion which couples to the tunneling induces the Kondo effect, provided the discrete vibrational energy compensates the singlet/triplet gap. We discuss also the possibility of tuning the phonon-induced Kondo tunneling by external magnetic field and the finite bias Kondo anomaly.

PACS: 85.65.+h; 73.23.Hk

Keywords: Kondo effect; Molecular transistor; Singlet/triplet transition

Various mechanisms of phonon-assisted tunneling through quantum dots and organometallic complexes (OMC) in Kondo regime are discussed in current literature [1]. Depending on the strength of vibronic coupling, either phonon satellites accompany the main Kondo peak or the phonon line shape modifies the Kondo-type zero bias anomaly (ZBA). In this work we describe an alternative scenario, where the Kondo tunneling between metallic electrodes via OMC is possible only due to virtual excitation of molecular vibrations in the process of single-electron transport. The Kondo channel opens provided the local vibration mode exists in OMC, which compensates the singlet–triplet (S–T) exchange gap Δ. Since many vibrational modes are available already for OMC of moderate size, this condition seems realistic enough.

Possible realization of vibration induced Kondo tunneling for OMC containing 3d ions was offered in Ref. [2]. In that case the desired S–T multiplet may be realized for configurations $3d^2(e^2)$ or $3d^8(t^6e^2)$ with e-orbital levels split by the ligand field of distorted cubic symmetry. Here we consider more symmetric case of 4f ions secluded in a cage with mirror symmetry plane perpendicular to the source–-drain direction: the cerocene molecule $Ce(C_8H_8)_2(4f^1\pi^3)$ where the magnetic Ce ion is sandwiched between two planes formed by molecular π orbitals [3] is a possible object. The ground and the first excited states state $|\Lambda\rangle$ of this molecule are the spin singlet $|^1A_{1g}\rangle$ and the triplet $|^3E_{2g}\rangle$.

One may write the Anderson Hamiltonian for an OMC with vibrating molecular shell in a tunnel contact with two metallic leads in the form $H = H_{OMC} + H_{tr} + H_{vib}$, where H_{OMC} stands for the isolated OMC,

$$H_{tr} = \sum_{k\sigma} \varepsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \hat{w}_Q \sum_{k\mu\sigma} \left(\tilde{d}^\dagger_{\mu\sigma} c_{k\sigma} + H.c. \right) \qquad (1)$$

describes electrons in the leads and the lead–dot tunneling, $H_{vib} = \hbar\Omega P^2/2$ describes oscillations in the shell. These vibrations are represented by a single harmonic mode with the frequency Ω and canonical variables P, Q. We assume that the electron–phonon interaction enters the model only through the tunnel matrix element $\hat{w}_Q$.

If H_{OMC} is represented only by its low-energy spin eigenstates $|\Lambda\rangle$, the effective spin Hamiltonian H_{SW} for the

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[1]Supported by the Heisenberg program of the DFG.

0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2006.10.801

Anderson Hamiltonian H may be derived by means of the Schrieffer–Wolff transformation [2]:

$$H_{\text{eff}} = \frac{1}{2}\Delta\mathbf{S}^2 + \hat{J}_S\mathbf{S}\cdot\mathbf{s} + \hat{J}_R\mathbf{R}\cdot\mathbf{s} + \frac{\hbar\Omega}{2}P^2. \tag{2}$$

The electron spin operator is given by the expansion $\mathbf{s} = \frac{1}{2}\sum_{kk'}\sum_{\sigma\sigma'} c^\dagger_{k\sigma}\boldsymbol{\tau}_{\sigma\sigma'}c_{k'\sigma'}$ where $\boldsymbol{\tau}$ is the Pauli vector. Two vectors describing transitions within the S–T multiplet are the spin 1 vector $\mathbf{S}$ and the vector $\mathbf{R}$ describing S–T transitions. These operators may be fermionized as $S^+ = \sqrt{2}(f^\dagger_0 f_{-1} + f^\dagger_1 f_0)$, $S^z = f^\dagger_1 f_1 - f^\dagger_{-1} f_{-1}$, $R^+ = \sqrt{2}(f^\dagger_1 f_s - f^\dagger_s f_{-1})$, $R^z = -(f^\dagger_0 f_s + f^\dagger_s f_0)$, where the indices $s, 0, \pm 1$ stand for singlet and three projections of triplet [4].

Phonon excitation enters the Kondo cotunneling Hamiltonian (2) via effective exchange constants $\hat{J}_\Lambda(Q) \approx |\hat{w}(Q)|^2/\varepsilon_\Lambda$, where ε_Λ are the corresponding electron ionization energies. Using fermionic representation for spin operators and expansion $Q = (b^\dagger + b)/\sqrt{2}$ for the displacement operator, one may describe the relevant processes for the phonon-assisted Kondo cotunneling by the diagram of Fig. 1a [2]. In accordance with Ref. [2], we retain only single-phonon processes for $\hat{J}_R(Q) = J_R + j_R Q$. The vertex corrections are calculated by means of analytical continuation of Matsubara-type diagrams from imaginary axis to the real frequency axis and summation of the parquet series [2]:

$$\gamma \sim (j_R)^2\rho\left[\frac{\log(D/\max[T, |\Delta - \hbar\Omega|])}{1 - J_S A\rho \log(D/\max[T, |\Delta - \hbar\Omega|])}\right]. \tag{3}$$

Here, $A \sim 1$ is a constant determined by spin algebra, D is the effective width of the electron conduction band and ρ is the density of states on the Fermi level. The Kondo temperature extracted from this equation reads $T_K \sim D\exp(-1/(A\rho J_S))$. One concludes from these calculations that the single-phonon processes are sufficient to compensate the energy of the S–T splitting and induce resonance tunneling through the OMC provided the local vibration mode with appropriate frequency satisfying the condition

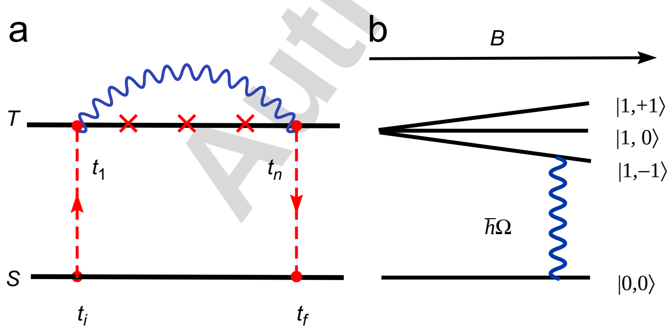

Fig. 1. (a) Phonon-assisted Kondo cotunneling process: virtual phonon absorption initiates a S–T transition, Kondo processes take place in an intermediate triplet state and the phonon is emitted in the end. (b) Magnetic field fine-tuning of the S–T Kondo resonance.

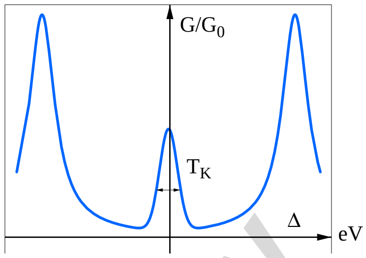

Fig. 2. Typical shape of the differential conductance as the function of finite bias eV. The first peak corresponds to ZBA while the second peak is attributed to the finite bias Kondo effect.

$$|\hbar\Omega - \Delta| \lesssim T_K \tag{4}$$

exists in the cage. One can expect in this case a significant enhancement of the tunnel conductance already at $T > T_K$ according to the law $G/G_0 \sim \ln^{-2}(T/T_K)$ [5], where G_0 is the conductance at unitarity limit $T \to 0$. We emphasize that in spite of the fact that the Kondo effect exists in our case only under phonon assistance, the Kondo temperature T_K is the same as in the usual Kondo effect. Since T_K is high enough ($\sim 10\,$K) in electro-migrated junction experiments with a OMC deposited between contacts [6,7], the effect predicted in this work seems to be easily observable. The crucial point is the existence of phonon satisfying condition (4) in a OMC with the S–T multiplet as a lowest spin excitation. One should note, however, that even if this condition is not exactly satisfied, one may tune the system by applying the magnetic field (Fig. 1b). Then the triplet is split, and only the level $E_{T,-1} = E_T - E_Z$ is involved in the phonon-induced Kondo tunneling (E_Z is the Zeeman energy). In this case Δ in (4) is substituted for $\Delta_Z = \Delta - E_Z$, and E_Z may be tuned to satisfy inequality (4). Thus, the vibration gives rise to a magnetic field induced Kondo effect at Zeeman energies which can be much smaller than Δ. The only difference is that in this case the effective spin of the OMC is one-half instead of one [8].

The differential conductance as a function of the bias eV is shown in Fig. 2. The central peak is suppressed at $eV \sim T_K$ due to the decoherence effects associated with the electrical current across the OMC. The conductance grows again at $eV \to \Delta$, due to non-equilibrium effects occurring when the resonance tunneling is restored at $eV = \Delta$ [9]. The width of the non-equilibrium peak is determined by the non-equilibrium Kondo temperature $T_K^{NEK} \sim (T_K)^2/D$. The Kondo-shuttling effect associated with the OMC centrum of mass motion, briefly discussed in Ref. [10], requires detailed analysis.

To conclude, we demonstrated that phonon emission/absorption can induce Kondo tunneling in OMC with even electron occupation and a spin singlet ground state, when the conventional Kondo effect is suppressed.

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