AIP Journal of Applied Physics

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D. Ciudad

Citation: Journal of Applied Physics **114**, 114508 (2013); doi: 10.1063/1.4821023
View online: http://dx.doi.org/10.1063/1.4821023
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JOURNAL OF APPLIED PHYSICS **114**, 114508 (2013)

Dependence of the conduction regimes of discontinuous magnetic tunnel junctions on clusters' volume and tunnel resistance

D. Ciudad[a)]
Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA and CIC nanoGUNE Consolider, Tolosa Hiribidea 76, 20018 Donostia-San Sebastian, Spain

(Received 24 June 2013; accepted 26 August 2013; published online 19 September 2013)

In a previous work [Phys. Rev. B **85**, 214408 (2012)], the conductance and the tunneling magnetoresistance in discontinuous high anisotropy magnetic tunnel junctions was experimentally studied. Different conduction regimes (sequential tunneling, co-tunneling, Kondo effect, and direct tunneling) and gradual transitions between them were found as a function of the temperature and the size of the clusters within the barrier. A simple theoretical model was suggested able to account for the experimental results even assuming no dispersion of the distribution of the size of the clusters. Here, we study, within this theoretical framework, the effect of the volume of the clusters within the barrier, and the effect of the thickness of the insulating barrier (or the tunnel resistance), on the transition between Kondo effect and co-tunneling regime. Clarifying the role of both parameters is of importance to understand and experimentally control the transition between the different conduction regimes. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4821023]

I. INTRODUCTION

Double magnetic tunnel junctions (DMTJs) with clusters within the insulator are one of the different possible approaches to develop single-electron spintronic devices. Conductive clusters electrically isolated have large charging energies which can give rise to the suppression of the charge transport or Coulomb blockade (CB) effect. When CB is large enough, single electron tunneling regime is possible.[1] However, the suppression is not perfect since it is still possible the simultaneous tunneling (or co-tunneling) of two electrons in and out of the cluster. CB and the Kondo effect can arise in quantum dots or nanocluster-based systems, such as DMTJs. While co-tunneling can give rise to an enhancement of the tunnel magnetoresistance (TMR), Kondo effect can suppress it.[1] Studying the transition between both conduction regimes is important to determine the limitations of future single electron spintronic devices on the one hand and to clarify the physical foundations of the co-existence of ferromagnetism and the Kondo effect on the other.[2,3]

Our work has been focused on DMTJs with high magnetic anisotropy $Co_{75}Pt_{25}$ nanoclusters embedded within the barrier. We experimentally found a temperature-tunned gradual transition between co-tunneling and Kondo, instead of the sharp crossover as previously reported in $Co_{70}Fe_{30}$ clusters.[2,3] This transition was reflected both on the conductivity and the TMR.

To explain these experimental results, a simple theoretical model was developed, similar to those used to explain the TMR in junctions with impurities in the barrier. In these works, three different conduction paths were considered: direct, through non-magnetic impurities, and through magnetic impurities. The last term was: (1) a semi-empirical expression introduced to take into account the spin flips the magnetic impurities were experimentally found to produce;[4–6] or (2) directly taken equal to the spin-exchange scattering found in Appelbaum and Anderson perturbation models.[7–9] We experimentally found that such spin flips are due to Kondo effect, a zero bias anomaly in DMTJs due to the contribution to the tunneling current of localized states within the barrier, validating the use of the spin-exchange term to explain the results of MTJs with magnetic impurities.[10] Notice that both Appelbaum and Anderson models were originally developed to explain the physical origin of Kondo effect.

Kondo effect increases the conductance while reducing the TMR in MTJs with barriers having magnetic clusters within them or with doped barriers with paramagnetic materials.[2,11,12] In co-tunneling regime, MTJs show the opposite behavior, an enhancement of the TMR, and a reduction of the conductivity. Therefore, the competition between Kondo effect and co-tunneling must be shown up both in the TMR and the conductivity.

Here, we explore the effect of the thickness of the barrier and the volume of the clusters in the transition between Kondo and co-tunneling regime within the frame of our theoretical model.

II. THEORETICAL MODEL

Let us briefly have an overlook to our theoretical model.[2] Different conduction channels are considered to compete and to contribute to the total conductance. The total TMR is due to the contribution of these different conduction channels.

To simplify, we will consider here the direct tunneling between the electrodes being negligible. In that case, according to the model, the conductivity σ is

$$\sigma = \sigma_E + \sigma_K. \quad (1)$$

[a)]Electronic addresses: dciudad@mit.edu and dciudad@nanogune.eu

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Here, σ_E the elastic conductivity through the clusters without spin flips, and σ_K the conductivity through the clusters showing Kondo effect, i.e., spin flips.

The Kondo conductivity for a broad range of temperatures and in particular for temperatures close to T_K is given by[13]

$$\sigma_K = \sigma_0 \left(\frac{T_{K2}^2}{T^2 + T_{K2}^2} \right)^S, \tag{2}$$

where

$$T_{K2} = \frac{T_K}{\sqrt{2^{1/S} - 1}}, \tag{3}$$

and, for symmetric barriers,

$$\sigma_0 \leq (2e^2)/h, \tag{4}$$

where e is the charge of the electron and h the Planck's constant. The parameter S takes the value 0.2 for temperatures close to T_K.[13]

The elastic term σ_E depends on the conduction regime (co-tunneling or sequential tunneling). Since we are just interested here on the gradual transition between Kondo effect and co-tunneling, we will simplify σ_E, neglecting the contribution of the sequential tunneling, and taken into account the co-tunneling term only (σ_{cot}). This approximation is valid for the limit ($k_B T \ll E_c$). Thus,

$$\sigma_E = \sigma_{cot} = \frac{2h}{3e^2} \frac{1}{R_T^2} \left(\frac{k_B T}{E_c} \right)^2, \tag{5}$$

being E_c the charging energy and R_T the tunnel resistance between one electrode and the clusters.

The different conduction channels have their own contribution to the total TMR. Assuming a perfect suppression of the TMR, when Kondo effect arises[2]

$$\mathrm{TMR} = \frac{\sigma_E}{\sigma} \mathrm{TMR}_E. \tag{6}$$

TMR_E is the elastic tunneling through the clusters. Since only co-tunneling is being taken into account, $\mathrm{TMR}_E = \mathrm{TMR}_{cot}$, where TMR_{cot} is the contribution to TMR due to the co-tunneling conduction. In a pure co-tunneling regime, TMR does not depend on temperature.

III. EFFECT OF THE VOLUME OF THE CLUSTERS

In the model, there are four different parameters: R_T, σ_0, E_c, and T_K. Let us consider first how the volume of the clusters affects the transition between Kondo dominated conductance and co-tunneling regime.

R_T mainly depends on the nature and the thickness of the insulating barrier between the electrode and the clusters (t). In Ref. 2, for DMTJs with $Co_{75}Pt_{25}$ clusters within a $t = 1.1$nm alumina barrier, R_T was found to be $4k\Omega$.

σ_0 is a function of maximum conductance in Kondo regime and the site occupancy.[2,13] In $Co_{75}Pt_{25}$ clusters, with $\sigma_0 \approx 10^{-3}(2e^2)/h$ the predicted gradual transition between Kondo and co-tunneling regime happens in the same range of temperatures which was experimentally found.[2] Since $\sigma_0 \ll (2e^2)/h$ the system is not really in a pure Kondo regime but in the mixed-valence regime. The small value of σ_0 is due to the relatively big size of the clusters we are exploring, making the localized states being close to the Fermi energy of the electrodes.[13] In this case, increasing the size of the clusters does not produce big changes on these states and σ_0 can be considered independent on V. For an analysis on the dependence of T_K and σ_0 on the energy of the localized state, see Ref. 13.

E_c and T_K depend on the volume of the clusters V within the barrier. The charging energy E_c can be roughly evaluated as[2]

$$\mathrm{E_c} = \frac{e^2}{4\pi\epsilon\epsilon_0} \sqrt[3]{\frac{1}{12}\frac{\pi}{V}}. \tag{7}$$

ϵ_0 is the vacuum permittivity and ϵ the relative permittivity of the insulating barrier. For Al_2O_3 $\epsilon = 8$.[2]

The dependence of T_K with the number of atoms (N_A) in ferromagnetic clusters is discussed in Ref. 14. Since $V \propto N_A$, it turns out that

$$\mathrm{T_K} = a\sqrt{\frac{1}{V}} e^{-V}. \tag{8}$$

The proportionality constant a can be evaluated from experimental data. For $Co_{75}Pt_{25}$ clusters with a volume $V = 15\mathrm{nm}^3$, we found $T_K \approx 10$K.[2]

Using the previous values for the different parameters, Figs. 1(a)–1(c) show the conductivity σ, the contribution of σ_K to the total conductance, and the TMR predicted by the theoretical model, as a function of V and T. The volume of the clusters V has been varied from 10 to 25 nm^3 which are the values experimentally found when depositing $Co_{75}Pt_{25}$ layers on alumina with nominal thicknesses of 0.5 and 1.1 nm, respectively.[2] Within this range, V seems to have little effect on the transition between Kondo and co-tunneling when varying the temperature. The range of temperatures predicted for the transition is similar to those experimentally found.[2]

Let us consider again the effect of V on the transition, but increasing R_T from 4 to $16k\Omega$ assuming no change on the other parameters, see Figs. 1(d)–1(f). This change reduces σ_E allowing the increase on the conductance due to Kondo effect be apparent at low temperature (<4 K). In addition, it makes the transition on the conduction regimes smoother since it broadens the span of temperatures where it takes place. On the other hand, the effect of V is also small in this case.

Comparing Figs. 1(a) with 1(d), 1(b) with 1(e) and 1(c) with 1(f), it is clear that the change on R_T from 4 to $16k\Omega$

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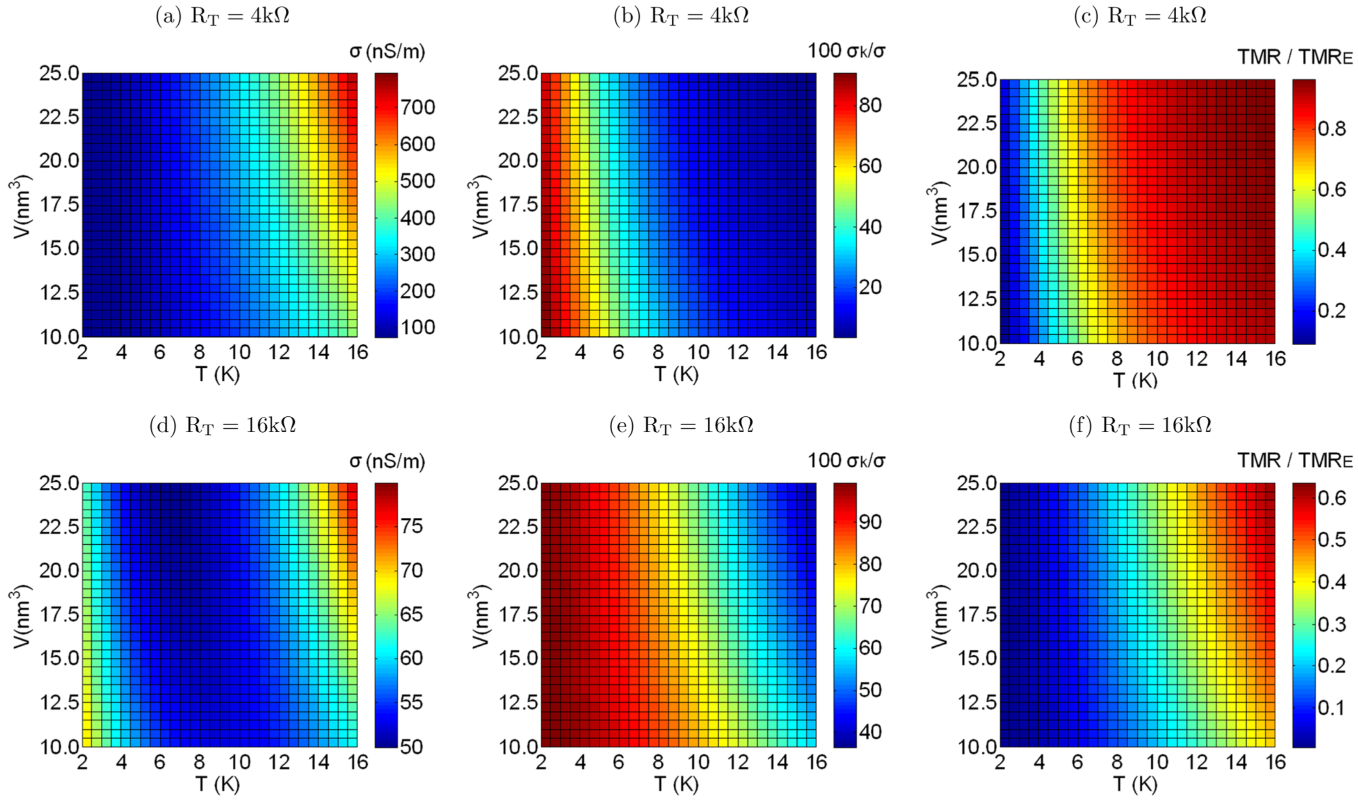

FIG. 1. Magnetotransport parameters as a function of temperature and the volume of the cluster within the barrier, for two different values of the tunnel resistance R_T. For $R_T = 4k\Omega$ it is given the conductivity (a), the relative contribution of the Kondo conductance to the total conductance (b), and the magnetoresistance normalized by the contribution of the elastic conduction (c). The same parameters are shown in (d)–(f) for $R_T = 16k\Omega$.

has a much higher effect on σ and TMR than any change on V (disregarding any associated change on R_T).

IV. EFFECT OF THE THICKNESS OF THE BARRIER OR THE TUNNELING RESISTANCE

The dependence of the transition between Kondo effect and co-tunneling with the thickness of the barrier between one of the electrodes and the clusters (t) is included in the term R_T. From Simmon's tunneling equations, $R_T \propto t^2 e^{bt}$.[15] The parameter b depends on the barrier potential. Let us assume clusters with a fixed volume ($V = 15\text{nm}^3$). Now R_T will be the variable. Two different cases are analyzed, one with a constant value of T_K, the other assuming an exponential dependence of T_K with the barrier thickness.

Figs. 2(a)–2(c) show σ, the contribution to the total conductance of σ_K and TMR as a function of R_T and T. Here, a constant value for Kondo temperature is assumed ($T_K = 10\text{K}$). As found in the previous section, the change on R_T has a huge impact on the transition between the conduction regimes. Increasing R_T, the transition is highly broaden and it takes place at higher temperatures.

The minimum value for R_T considered is $4k\Omega$ since this is the experimental value we found for MTJs with an alumina barrier with thickness $t = 1.1\text{nm}$.[2] Smaller resistances can be achieved by reducing the thickness t. However, in addition to the technical problem it could suppose, it could make the coupling energy (Γ) between the electronic states of the clusters and those on the electrodes large enough to destroy the quantization of charge and energy even at very low temperatures.[16] Note that the existence of Kondo effect in MTJs is itself a manifestation of such a strong coupling between the ferromagnetic electrodes and the clusters through the insulating barriers.[17]

The maximum value for R_T we considered is $16k\Omega$, just four times higher than the minimum one. This value can be achieved by slightly increasing the thickness of the tunneling barrier, since R_T depends exponentially on t. It allows us to consider the parameters involved in the Kondo term to be constant. This is supported by the experimental results by Yang *et al.*[17] In this work, the thickness of MgO barriers needs to be largely increased from 2.8nm to 3.6nm in order to avoid the characteristic double peak on the dynamic conductance due to Kondo.

These experimental evidences show that taking T_K constant for the values of R_T considered is reasonable. However, let us assume now that a strong dependence happens. Let us consider the extreme case of an exponential decay of the coupling energy Γ with the thickness of the barrier. While ($k_B T_K < \Gamma$) is satisfied, an exponential decay with t is reasonable to be considered for T_K.[16] It follows from here that $T_K \propto R_T^{-1}$. The proportionality constant is found from the experimental data ($T_K = 10\text{K}$ and $R_T = 4k\Omega$ for clusters with volume $V = 15\text{nm}^3$). Using this dependence for T_K, the predictions of the model are shown in Figs. 2(d)–2(f). Comparing these graphs with Figs. 2(a) and 2(b), it is clear

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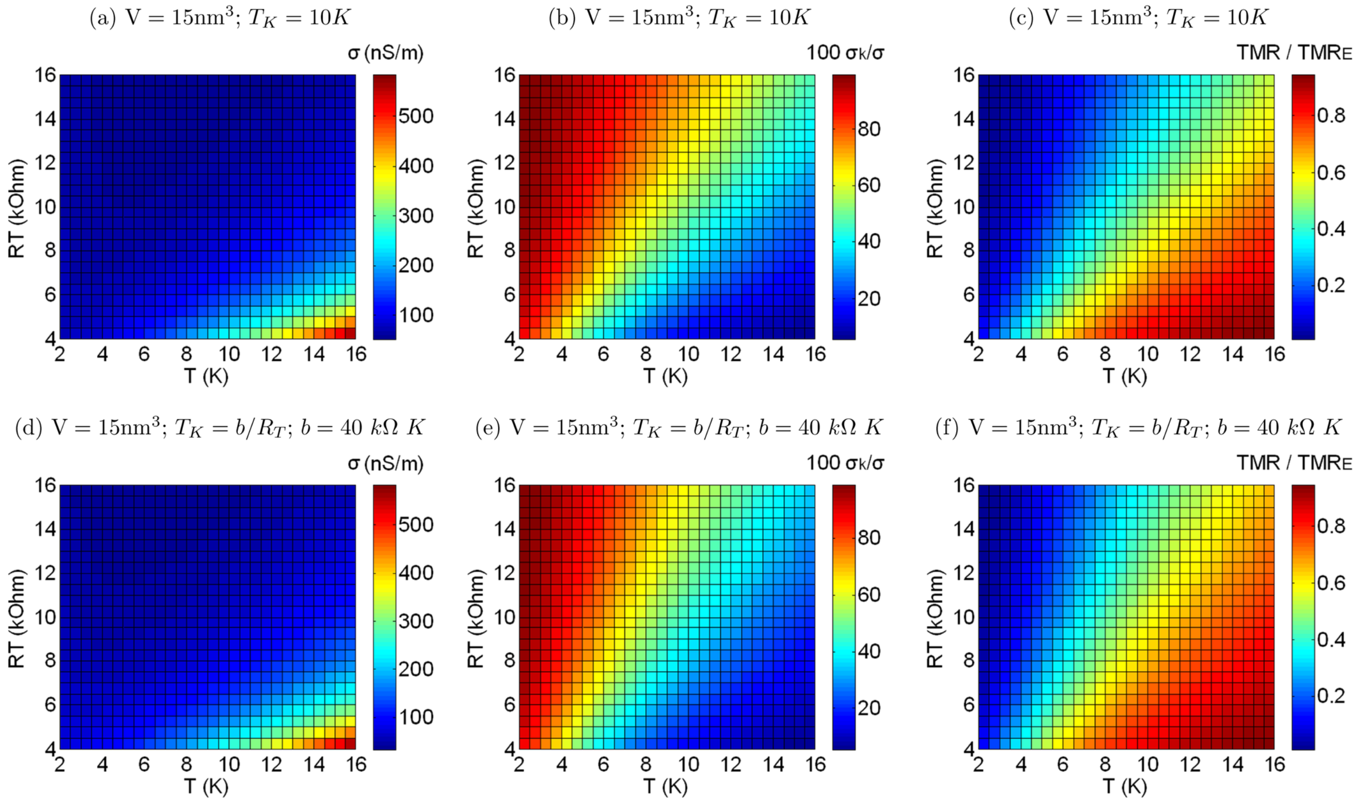

FIG. 2. Magnetotransport parameters as a function of temperature and the tunnel resistance. (a)–(c) show the conductivity, the relative contribution of the Kondo conductance to the total conductance, and the magnetoresistance normalized by the contribution of the elastic conduction, considering a constant value of the Kondo temperature T_K. The same parameters are shown in (d)–(f) supposing an exponential dependence with the barrier thickness.

that the change on T_K with R_T is an small correction at least on the span of values for the different parameters considered.

V. CONCLUSION

We have developed a theoretical model that predicts a gradual transition tunned by temperature between Kondo and co-tunneling dominated regimes in DMTJs, even for no-dispersion of the cluster's size. This transition affects both the conductivity and the TMR. As previously shown, this is good agreement to our experimental findings.[2] Here, we have studied the effect of the volume of the clusters and the thickness of the barrier or tunnel resistance, which are parameters that are easily controlled experimentally. According to the model, the change on the volume disregarding any related change R_T has little effect on the transition, whereas the change on the tunnel resistance largely modifies the span of temperatures where it takes place.

The experimental differences between $Co_{70}Fe_{30}$ and $Co_{75}Pt_{25}$ clusters (well defined crossovers vs. gradual transitions) are not due to the cluster's size dispersion since it was found to be similar in both cases. This conclusion is supported also by the little effect on volume theoretically found here. In Ref. 2, we suggested that these experimental differences could be due to different values of σ_0 and T_K coming from differences on magnetic anisotropy. However, here, we show that these differences could simply be due to the different values of the tunnel resistance coming from the different nature and thickness of the insulating barrier.

ACKNOWLEDGMENTS

The author wishes to thank Professor C. H. Marrows' supervision and kind support during the author's postdoctoral research stay at the University of Leeds. Support from the Spanish Ministry of Science and Innovation through postdoctoral Grant No. 2008-0352, and a Marie Curie International Outgoing Fellowship within 7th Framework Programme is acknowledged.

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