23. E.W. Russell, L.B. Weiss, F.I. Navetta, R.E. Koeppe and O.S. Andersen, Biophys.J., 49,6673(1986).
24. F. Heitz, N. Daumas, N. Van Mau, R. Lazaro, Y. Trudelle, C. Etchebest and A. Pullman, "Transport Through Membranes: Carriers, Channels and Pumps", A. Pullman, J. Jortner and B. Pullman, eds., Kluwer Academic Publishers, London, 1988, p147.
25. R.E. Koeppe, O.S. Andersen and J.-L. Mazet, 8 International Biophysics Congress, 273(1984).
26. R.E. Koeppe, O.S. Andersen and A.K. Maddock, "Transport Through Membranes: Carriers, Channels and Pumps", A. Pullman, J. Jortner and B. Pullman eds., Kluwer Academic Publishers, London, 1988, p133.
27. C. Etchebest and A. Pullman, J.Biomol.Structure and Dynamics, 2,859(1985).
28. J.A. Killian, J.W. Timmermans, S. Keur and B. Dekruijff, Biochim.Biophys.Acta, 820,154(1985).
29. P. Cavatorta, A. Spisni, E. Casali, L. Lindner, L. Masotti and D.W Urry, Biochim.Biophys. Acta, 689,113(1982).
30. L. Masotti, P. Cavatorta, G. Sartor, E. Casali and A.G. Szabo, Biochim.Biophys. Acta, 862,256(1986).

# THE KONDO EFFECT IN ELECTRON SPIN RESONANCE IN MAGNETIC ALLOYS

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As the temperature decreases (T remains above the Kondo temperature $T_k$) effeciency of the relaxation processes in spin subsystems of localized moments and conduction electrons in dilute magnetic alloys, caused by the exchange interaction, increases. This leads to emergence of the coupled motion of magnetic moments of impurities and conduction electrons in the system. The aim of the present work is the microscopic study of the influence of the Kondo effect on the EPR parameters of dilute magnetic alloys, having taken into consideration the dynamic nature of the exchange coupling between localized magnetic moments and conduction electrons.

1. The Hamiltonian, which describes the system of localized moments and conduction electrons in the external constant magnetic field, can be written in the following form [1,2]

$$H = H_0 + H_{es} + H_{eL} \qquad (1)$$

Here, $H_0$ is the Hamiltonian of localized spins and conduction electrons in the constant magnetic field; $H_{es}$ and $H_{eL}$ determine the interaction of conduction electrons with magnetic and nonmagnetic impurities, respectively.

The Feynman diagram techniques, the methods of the dynamical renormalization groups and the Zubarev's nonequilibrium statistical operator method are used in calculations and a pseudofermion representation of the localized spin operators is introduced.

To evaluate the total response of the system the Bethe-Salpeter equations for the vertex parts of the two-particle Green functions, which define the dynamic transverse susceptibilities of localized moments and conduction electrons, are constructed to within the third order terms in the exchange interaction constant J, second order terms in the impurity potential and spin-orbit scattering amplitudes V and $V_{so}$, respectively, and first order terms in concentrations of impurities [3]. All terms in the equations are expressed through "dressed" single particle Green functions, calculated to within the same order terms in the interactions. For high ( $kT \gg \omega_s, \omega$ ) and low ( $kT \ll \omega_s, \omega$ ) temperatures the integral vertex equations are reduced to the following coupled system of equations for the dynamic transverse susceptibilities of localized moments $\chi_s(\omega)$ and conduction electrons $\chi_e(\omega)$

$$\begin{aligned} a_s \chi_s(\omega) + b_e \chi_e(\omega) &= c_s \ , \\ b_s \chi_s(\omega) + a_e \chi_e(\omega) &= c_e \ , \end{aligned} \qquad (2)$$

the parameters $a_i, b_i, c_i$ ( $i = s, e$ ) are defined by the expressions:

$$a_i = \omega_i' - \omega - \theta_i \Sigma_{ij} - \Sigma_{iL},$$

$$b_i = \lambda \chi_j (\Sigma_{jL} - \omega_j) + g_i \theta_i \Sigma_{ij} / g_j \ ,$$

$$c_i = \chi_i(\omega'_i - \Sigma_{ij} - \Sigma_{iL}) + g_i \chi_j \Sigma_{ji}/g_j,$$

where

$$\omega'_i = \omega_i(1+\lambda\chi_j), \quad \theta_i = 1+\lambda\chi_i g_j/g_i ,$$

$$\chi_s = (C_M g_s^2/2\omega_s)\,\mathrm{th}(\beta\omega_s/2), \quad \chi_e = g_e^2 d/2$$

$$\Sigma_{ij} = \mathrm{Re}\Sigma_{ij} + i\mathrm{Im}\Sigma_{ij}, \quad (ij = se, es, eL),$$

$$\mathrm{Im}\Sigma_{eL} = (2/3)\pi d|V_{sc}|^2 C_{sc} \int d\Omega \sin\theta + Dq^2,$$

$$\tau_p^{-1} = 2\pi d C_p |V|^2, \quad D = v_F^2 \tau_p/3, \quad \mathrm{Re}\Sigma_{eL} = 0$$

$C_M$, $C_p$, $C_{sc}$ are concentrations of localized spins, nonmagnetic impurities with spin-independent and spin-orbit potentials, respectively; 2D is the bandwidth; d is the constant density of states. The imaginary parts $\Sigma_{ij}$ $(ij = se, es, eL)$ have the meanings of transverse relaxation rates of the spin magnetizations of localized moments and conduction electrons, caused by the exchange interaction and spin-orbit scattering of conduction electrons from nonmagnetic impurities. The real parts $\Sigma_{ij}$ determine shifts of the resonanse frequencies of localized moments and conduction electrons.

The next higher-order corrections in the exchange interaction in the coupled Bethe-Salpeter equations for vertex parts of two-particle Green functions of localized moments and conduction electrons only redefine the kinetic coefficients and the form of the equations (2) is not changed [3]. Using the method of the dynamical renormalization groups [4], we get the following expressions for kinetic coefficients

$$\mathrm{Im}\Sigma_{se} = \begin{cases} \pi kT \ln^{-2}(T/T_K), & kT >> \omega_s, \\ \pi\omega_s \ln^{-2}(\omega_s/kT_K)/4, & kT << \omega_s, \end{cases} \quad (3)$$

$$\mathrm{Im}\Sigma_{es} = \begin{cases} \pi C_M/2d \ln^2(T/T_K), & kT >> \omega_s, \\ \pi C_M/4d \ln^2(\omega_s/kT_K), & kT << \omega_s, \end{cases} \quad (4)$$

$$\mathrm{Re}\Sigma_{se} = (g_s/g_e)\,\omega_s \lambda \chi_e \cdot$$

$$\cdot[(2dJ\ln(kT_K/Y))^{-1} - 1], \quad (5)$$

$$\mathrm{Re}\Sigma_{es} = \omega_e \lambda \chi_s \cdot$$

$$\cdot[(2dJ\ln(kT_K/Y))^{-1} - 1], \quad (6)$$

$$kT_K = D\exp(1/2dJ), \quad (7)$$

where $Y = \max\{kT, \omega_s\}$.

Using the Zubarev's nonequilibrium statistical operator method [2], we obtain the system of coupled equations for the dynamic susceptibilities of localized moments and conduction electrons, which is valid for all temperatures and external frequencies and which coincides with (2), with the kinetic coefficients $\Sigma_{se}$ and $\Sigma_{es}$, calculated to the third order in J:

$$\mathrm{Re}\Sigma_{se} = 2(dJ)^2 \cdot$$

$$[\omega \ln|D/\omega| - (\omega_s - \omega)\ln|D/\omega_s - \omega|], \quad (8)$$

$$\mathrm{Im}\Sigma_{se} = \pi(dJ)^2 \cdot$$

$$[(\omega_s - \omega)\,\mathrm{cth}(\omega_s - \omega/2T)K + \omega\,\mathrm{cth}(\omega/2T)M], \quad (9)$$

$$\mathrm{Re}\Sigma_{es} = 2(dJ^2/C_M)\,\mathrm{th}(\omega_s/4T)$$

$$[\omega \ln|D/\omega| - (\omega_s - \omega)\ln|D/\omega_s - \omega|], \quad (10)$$

$$\mathrm{Im}\Sigma_{es} = \pi dJ^2 C_M \,\mathrm{th}(\omega_s/2T) \cdot$$

$$[(\omega_s - \omega)\,\mathrm{cth}(\omega_s - \omega/2T)K + \omega\,\mathrm{cth}(\omega/2T)M], \quad (11)$$

$$K = 1 - 4dJ(\omega_s \ln|D/\omega_s| - \omega \ln D/\omega)/(\omega_s - \omega),$$

$$M = 1 - 4dJ[\ln|D/\omega_s - \omega| - (\omega_s/\omega)\ln|\omega/\omega_s - \omega|].$$

Calculations of the kinetic coefficients on the basis of the dynamical renormalization groups' method [4] lead to the expressions (3)-(6) for the case $\omega = \omega_s$.

2. From equations (2) we obtain the following expression for the total dynamic susceptibility $\chi(\omega)$, which determines the response of the system of magnetic impurities and conduction electrons in dilute magnetic alloys

$$\chi(\omega) = (c_s a_e - c_e b_e + c_e a_s - c_s b_s)$$

$$(a_s a_e - b_s b_e)^{-1}, \quad (12)$$

The poles of the summary dynamic susceptibility $\omega_1$ and $\omega_2$ determine the coupled spin modes of magnetic impurities and conduction electrons. The strong bottleneck regime for conduction electrons in the

system is defined by the condition

$$Im(\theta_s\Sigma_{se}+\theta_e\Sigma_{es}) >> Im\Sigma_{eL}, |\omega_s' - \omega_e'|$$

In these conditions Kondo anomalies in EPR parameters for dilute magnetic alloys are fully suppressed. This is due to the fact that under these conditions the magnetic impurities' and conduction electrons' spins are tightly coupled to each other, and magnetization can only "leak" to lattice [3]. We shall now consider the intermediate bottleneck EPR regime in dilute magnetic alloys, which is characterized by the inequalities

$$|\omega_s' - \omega_e'| > Im(\theta_s\Sigma_{se}+\theta_e\Sigma_{es}) > Im\Sigma_{eL}$$

The poles of the summary dynamic susceptibility are equal to

$$Re\,\omega_1 = \omega_e' - Re(\theta_s\Sigma_{se}+\theta_e\Sigma_{es}) + (1-Q)Re\Sigma_{se}, \quad (13)$$

$$Im\omega_1 = Im\Sigma_{eL} + Im(\theta_s\Sigma_{se}+\theta_e\Sigma_{es}) - (1-Q)Im\Sigma_{se}, \quad (14)$$

$$Re\,\omega_2 = \omega_s' - Q\,Re\Sigma_{se}, \quad (15)$$

$$Im\,\omega_2 = QIm\Sigma_{se}, \quad (16)$$

where

$$Q=1-(g_e/g_s)\omega_s'\lambda\chi_s(\theta_s+\theta_e)/(\omega_e'-\omega_s')$$

According to (3)-(7),(13)-(16) the expressions for real and imaginary parts of the pole $\omega_2(\omega_1)$ which determine the effective resonance frequency and the effective linewidth of EPR on localized moments (conduction electrons) with the corrections, caused by the partial conservation of the coupled motion of magnetic moments of impurities and conduction electrons, contain the Kondo anomaly terms. According to (15),(16) as a consequence of the partial conservation of the coupled motion of magnetic moments of impurities and conduction electrons the effective g-value shift and the effective linewidth of EPR on localized moments depend on concentration of magnetic impurities. In the system of localized moments and conduction electrons the electron bottleneck regime is absent, if the following relation holds

$$Im\Sigma_{eL} >> Im(\theta_s\Sigma_{se}+\theta_e\Sigma_{es}), |\omega_s' - \omega_e'|$$

The collective oscillation of the spin density of localized moments and conduction electrons in the system does not take place in these conditions and the response of the localized spins and conduction electrons can therefore be considered separately. The Kondo anomalies are independently and fully exhibited in the parameters of EPR on localized moments as well as on conduction electrons [3].

3. It is interesting to compare the obtained theoretical results with the experimental data on EPR in Au:Yb from [5]. To fit the experimental data on the temperature dependences of the g-value shift and the spin relaxation rate of localized moments in Au:Yb using formulas, which are obtained neglecting the dynamic coupling between the spin subsystems of the localized moments and conduction electrons, two substantially differing in values the Kondo temperatures: $T_K^s = 4\cdot10^{-8}$ K and $T_K^{rel} = 2\cdot10^{-12}$ K, were introduced in [5]. However, the analysis of the experimental results of EPR in Au:Yb in [5] shows that the effective g-value shift and the effective relaxation rate of localized moments depend on concentration of magnetic impurities, besides the spin relaxation rate of localized moments in [5] is higher at low temperatures for alloys with higher concentration of magnetic impurities and at high temperatures - in samples with lower concentrations. According to (15),(16), the intermediate electron bottleneck regime in the conditions of the experiment was present. Therefore the interpretation of the experimental results should be based on the formulas (15),(16). Calculations in accordance with (15),(16),(3), (5),(7) of the actual Kondo temperature, using the experimentally obtained $T_K^s$ and $T_K^{rel}$ for $g_e=2$; $g_s=3,423$; D=100 K [5]; 2dJ=-0,05; $\lambda\chi_s = -0,5$ give $T_K^{exp} = 2\cdot10^{-7}$ K. The obtained $T_K^{exp}$ coincides with

$T_K^{theor}$ calculated in accordance to (7) for the same values of parameters. Consequently there is no necessity to introduce two specific to fit the EPR experimental data the Kondo temperatures $T_K^{\varepsilon}$ and $T_K^{rel}$, respectively. The parameters of EPR in dilute magnetic alloys contain only one Kondo temperature, which enters in the expressions for static and dynamic characteristics of dilute magnetic alloys, studied by other methods.

ACKNOWLEDGEMENTS

The authors are grateful to B.I.Kochelaev for a fruitful discussion of the results and for valuable comments.

References

1.Barnes S.E., Zitkova - Wilcox J Phys.Rev.B7, 2163 (1973).
2.Fazleyev N.G. Sov J.Low Tem.Phys 6, 693 (1980).
3.Mironov G.I.,Fazleyev N.G. Sov. J.Low Tem.Phys. 9, 950 (1988).
4.Fowler M, Zawadowski A Solid St. Commun. 9, 471 (1971).
5.Spalden Y. et al. Phys.Rev. B28, 24 (1983).