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ABSTRACT

The interaction between electrons and TLS (two level system) is studied in the simplest model. Using scaling arguments it is shown that the motions of TLS and of the electron screening cloud may be strongly correlated in the groundstate. Different measurable quantities are estimated.

АННОТАЦИЯ

При помощи простейшей модели исследуется взаимодействие между электронами и двух-уровневой системой (TLS). С использованием выводов теории подобия показано, что в основном состоянии движение двух-уровневой системы и экранирующего электроны облака может быть сильно взаимосвязано. Дана оценка некоторых измеряемых величин.

KIVONAT

Az elektronok és a két-nivós rendszer (TLS) közötti kölcsönhatást tanulmányozzuk a legegyszerübb modellen. Skálamegfontolásokkal megmutatjuk, hogy a két-nivós rendszernek és az elektronokat leárnyékoló felhőnek a mozgása az alapállapotban erősen korrelált lehet. Különböző várható mennyiségekre adunk becslést.

INTRODUCTION AND HAMILTONIAN

In the last years the model of TLS has generally been accepted for the low energy excitation in glasses (1) and in metallic glasses (2). An intuitive model for interaction between TLS and conduction electrons has been suggested by Cochrane et al.(3) in order to explain resistivity minima observed in many metallic glasses (4). It has been pointed out first by Kondo (5) that the screening of the tunneling atoms by conduction electrons results in infrared divergencies (see ref.6). Kondo (7) has also suggested that the structure of the interaction in real space may be of importance. The recent studies of the low energy state is reviewed, where the tunneling atoms are closely followed by the electron screening cloud. The formation of this state of new type is very sensitive on the initial parameters and shows a strong resemblance with the Kondo state in dilute magnetic alloys. The most direct information on the interaction between TLS and electrons are provided by different ultrasound experiments (see e.g. ref. 8 and 9).

The TLS described by pseudospin in the usual way. The Hamiltonian is given in terms of Pauli operators σ^i_{TLS} $(i=x,y,z)$

$$H_o = \frac{1}{2} \left(\Delta\, \sigma^z_{TLS} + \Delta_o\, \sigma^x_{TLS} \right) \qquad 1$$

where Δ is the energy splitting and Δ_o is the tunneling rate between the potential wells, thus $\Delta_o \sim \hbar\omega_o e^{-\lambda}$ with the zero point energy $\hbar\omega_o$ in the well. This Hamiltonian can be diagonalized by a rotation around the y-axis and in the new representation $H_o = \frac{1}{2} E\, \bar{\sigma}^z_{TLS}$, where $E = \sqrt{\Delta^2 + \Delta_o^2}$ Furthermore, it is generally accepted for a glass that the distribution of energies E is homogeneous, thus $P(E)=P_o$.

The electron gas is considered like free electrons and the electron states are characterized by momentum k and energy $\varepsilon(k)$, futhermore, the Hamiltonian is $H_e = \sum_{ks} \varepsilon(k) a^+_{ks} a_{ks}$, where a_{ks} is the electron annihilation operator with spin s. The general form of the interaction Hamiltonian between electrons and TLS is

$$H_1 = \sum_{\substack{kk's \\ i=x,y,z}} a^+_{ks} V^i_{kk'} a_{k's} \sigma^i_{TLS} \qquad 2$$

where $V^i_{kk'}$ (i=x,y,z) are the coupling constants. It will be useful to write $V^i_{kk'}$ in the form $V^i_{kk'} = \sum_{\alpha\beta} f^*_\alpha(k)^+ V^i_{\alpha\beta} f_\beta(k')$, where the functions f_α form a complete set which depend only on the direction of $\vec{k}$ (e.g. spherical harmonics).

There are scattering processes, which are diagonal and off-diagonal in the TLS variables. V^z describes the difference of the two scattering amplitudes corresponding to the two positions of the TLS. The electron assisted tunneling is described by V^z and V^y, thus $V^x\, V^y \sim e^{-\lambda}$.

For the sake of simplicity let us consider the TLS as a single atom in the double well potential, and the electron is scattered by the atom in the s-wave channel only. This scattering is described by a pseudopotential U. The separation between the two positions is denoted by $\vec{d} \,||\, z$. According to Black et al.(10) and Kondo (7) V^z is

$$V^z_{kk'} = \frac{1}{2}\, i(k_z - k'_z)\, dvU \qquad 3$$

where v is the atomic volume.

In the electron assisted tunneling the effective potential barrier for the TLS atom is $V(x) = V_B(x) + U\delta\rho(x)$, where V_B is the potential without electron density fluctuation $\delta\rho(x)$. In order to get the rate of the electron assisted process the functional derivative of the tunneling rate must be taken with respect $\delta\rho(x)$. The result is $(k_z - k'_z)^2 d^2 \lambda v U \rho_o \Delta_o / V_B$. The factor $(k_z - k'_z) d^2$ is due to the fact, that V(x) must be measured with respect the average of the potential minima, because a homogeneous fluctuation is not effective. Similar expression was proposed by Kondo (7). As the assisted tunneling may contribute to V^x and V^y as well, the structure of V^x and V^z must be studied. According to Vladár (11) the consequences of Hermiticity and time reversal symmetry of the Hamilton operator are $V^i_{kk'} = V^{i*}_{k'k}$ furthermore, $V^i_{kk'} = V^{i*}_{-k-k'}$ (i=x,z), and $V^y_{kk'} = -V^{y*}_{-k-k'}$, respectively. From these follows for the bare Hamiltonian studied, that

$$V^x_{kk'} \sim (k_z - k'_z)^2 d^2 \lambda v U \,\frac{\Delta_o}{V_B} \quad \text{and} \quad V^y_{kk'} = o \qquad 4$$

and $V^x/V^z \sim k_F d\lambda\, \Delta_o/V_B$. This ratio can be estimated as $V^x/V^z \sim 10^{-3}$-10^{-4} for a large amount of TLS by using the following values λ=6, d=0.3–0.5 Å^{-1} k_F= 1Å^{-1}, Δ_o=1K°, V_B=0.1eV.

Kondo (7) has pointed out that the Hamiltonian studied here results many different logarithmic corrections, furthermore, in order to get logarithmic correction to the electrical resistivity the momentum dependence given by eqs. 3-4 must be kept. The typical logarithmic term is $\log(D/\max(E,T))$, where D is the width of the conduction band ($D\sim 10$ eV) and T is the temperature. In this case simple weak coupling perturbation theory does not work, because of the big logarithmic factors. To have a clearer insight into such problem scaling argument can be applied to eliminate the large logarithmic corrections.The original problem with coupling V^i and with cutoff D is replaced by a similar problem with $V^{i'}$ and with a smaller cutoff D' in such a way that those phenomena are not modified in which electrons nearby the Fermi surface are involved. The goal of the procedure is to transfer the role of logarithmic terms to the strengths of the new couplings. This mapping can be cast into the form of a differential equation as

$$\frac{\partial V_{kk'}^{\ s}}{\partial \ln x} = 2\, i\, \rho_o \int \frac{dS_{\bar{k}}}{S_F} \left(V_{k\bar{k}}^{\ i}\, V_{\bar{k}k'}^{\ j}\right) \varepsilon^{ijs} \qquad 5$$

where $x = (D'/D)$ and the surface integral is taken along the Fermi surface with area S_F and ε^{ijk} is the Levy Civita symbol. This equation is the first order scaling equation as only the second order term is kept on the left hand side. It is important to notice, that the momentum dependence of the couplings may have crucial importance. Two cases must be distinguished (12)

(i) Commutative model, where $\int dS_{\bar{k}}(V_{k\bar{k}}^{\ i}\, V_{\bar{k}k'}^{\ j})\, \varepsilon^{ijs} = 0$ and therefore the couplings are invariant in the scaling of first order.

(ii) Non-commutative model, where $\int dS_{\bar{k}}(V_{k\bar{k}}^{\ i}\, V_{\bar{k}k'}^{\ j})\, \varepsilon^{ijs} \neq 0$ and the couplings are changed. If we start only with two couplings (e.g. V^x and V^z) the third one V^y is generated.

COMMUTATIVE MODEL ($V^x=0$ AND $V^y=0$)

Here it is assumed that only the coupling V^z is relevant. In case of a rotation around the y-axis there are formally two new couplings $\bar{V}^x$ and $\bar{V}^z$ ($V^{\perp}$ and $V^{||}$ in ref. 2), but they commute. The first scaling arguments were proposed by Black and Gyorffy (6). Recently, it has been reinvestigated in collaboration by Black, Gyorffy, Vladár and the author (13). In second order scaling the main results are for the scaled quantities $\bar{V}^{x'}$, $\bar{V}^{y'}$ and E'

$$(\bar{V}^{x'})^2+(\bar{V}^{z'})^2 = (V^z)^2 = \text{inv} \quad \text{and} \quad \frac{\bar{V}^{x'}}{\bar{V}^{z'}} = \frac{\bar{V}^x}{\bar{V}^z}\left(\frac{D'}{D}\right)^{(V^z)^2} \qquad 6$$

thus the coupling strength is invariant and the off-diagonal coupling $\bar{V}^{x'}$ is screened. The important features of this scaling theory are the following. Let us start in a rotated frame, thus $\Delta=E$ and $\Delta_o=0$. There are two steps in the scaling transformation (i) by changing $D\to D'$ the couplings are not changed, but Δ and Δ_o are modified. (ii) rotation around the y-axis to eliminate Δ_o. The main effect is in the screening of the energy splitting E of the TLS, as

$$(E'/E)^2 = [\ 1+(\bar{V}^{x'}/\bar{V}^{z'})^2\ ]\ /\ [\ 1+(\bar{V}^x/\bar{V}^z)^2\ ] \qquad 7$$

It has been shown (13), that the electrical resistivity does not change in the leading and next to the leading logarithmic order (e.g. $V^3\log$, $V^4\log$, $V^4\log^2$ terms do not exist). Smaller logarithmic terms like $\log(E/T)$ has not been ruled out.

NON-COMMUTATIVE MODEL

It will be seen, that if $V^z\rho_o \gtrsim 0.25$ then V^x is of importance, even if $V^x \ll V^z$ (ρ_o is the conduction electron density of states for one spin direction). The first scaling treatment of the non-commutative model has been given by Zawadowski (14), where only $\alpha,\beta=1,2$ kept in the matrix $V^i_{\alpha\beta}$. This assumption has been justified by Vladár (11). The first order scaling equation (see eq. 5) is in the matrix form as

$$\frac{\partial\, V^s_{\alpha\beta}}{\partial\, \ell n\, x} = 2\, i\, \rho_o \sum_\lambda V^i_{\alpha\gamma}\, V^j_{\gamma\beta}\, \varepsilon^{ijs} \qquad 8$$

In the region V^x, $V^y \ll V^z$ one can write a second order differential equation for V^x and V^y

$$\frac{\partial^2 V^i_{\alpha\beta}}{\partial\, \ell n x^2} = 4\, \rho_o^{\,2}\, [\, V^z_{\beta\beta} - V^z_{\alpha\alpha}]^2\, V^i_{\alpha\beta} \qquad (\ i = x,\ y\ ) \qquad 9$$

where the basis $f_\alpha(k)$ is chosen in such a way that V^z is diagonal. Considering the order of magnitude of V^x and V^y one should keep only that pair of α,β for which $(V^z_{\alpha\alpha} - V^z_{\beta\beta})^2$ is the largest. ($f_1 \sim Y^o_o(k) + i\, Y^o_1(k)$ and $f_2 \sim Y^o_o(k) - i\, Y^o_1(k)$ with m=0, where the quantum number m is related to the rotation around the z-axis, and Y^m_ℓ is the spherical function).

In the following we keep f_1 and f_2. This model has been discussed in great detail using first order scaling and a representation in which $V^i_{\alpha\beta} = \Sigma\, V^i_j\, \sigma^j_{\alpha\beta}$ (summation goes with j=x,y,z). The scaling given by eq. 8 leads to three coupling vectors $\vec{V}^i = (V^i_x\ V^i_y\ V^i_z)$ which are perpendicular and their lengths go to infinity, but their ratios become the same. Thus the model scales to an isotropic model. After long enough scaling and in appropriate base ($V^i_{\alpha\beta} = V\sigma^i_{\alpha\beta}$) a single coupling V remains. Thus the scaling leads to the following Hamiltonian

$$H_1 = V \sum_s \int dkdk'\, A^+_{\alpha s}(k)\, \vec{\sigma}_{\alpha\beta}\, A_{\beta s}(k')\, \vec{\sigma}_{TLS} \qquad 10$$

where $A_{\alpha s}(k) = \int dS_{\vec{k}}\, f_\alpha(\vec{k})\, a_{ks}$. The scaled Hamiltonian has the form of an antiferromagnetic Kondo Hamiltonian where the conduction electron magnetisation interacts with a localized spin with $S=1/2$. This result can be interpreted in the following way. In the magnetic spin problem the order of the spin-flip and non-spin-flip processes are of importance and the conduction electron spin polarization keeps a memory of that order. In the present case, the TLS has a pseudospin variable,and the conduction electrons have a distribution in the real space (angular dependent Friedel oscillation). In the non-commutative model the memory on the order of different scattering processes is in the conduction electron density polarization. In the strong coupling region the atoms of the TLS and the screening charge density are moving in strong correlation. This correlation shows up in the scaled Hamiltonian as the conservation of the total pseudospin (sum of pseudospins of the TLS and of the electron cloud). Using the analogy of the magnetic Kondo problem one can conclude that at low enough temperature a bound state or resonant state is formed with total pseudospin zero.

The second order scaling eqs. (15) are the following:

$$x \frac{\partial}{\partial x} v^i(x) = - 4\, v^j(x)\, v^k(x) + 8\, v^i(x)\, (v^j(x)^2 + v^k(x)^2) \qquad 11$$

with $i \neq j \neq k$ and

$$x \frac{\partial}{\partial x} \ln \Delta_o(x) = 8\, (\, v^z(x)^2 + v^y(x)^2\,) \qquad 12$$

where $x=D/D'$ and $v^i= V^i \rho_o$. These equations are applicable as far as $D' \gtrsim \max(E,T)$ and for the sake of simplicity the symmetric model is considered (11, 15).

The higher order terms are negligible if $v^i=V^i \rho_o \lesssim 0.2$. The fixed point $v^{x*}=v^{y*}=v^{z*}=1/4$ of eq.11 is already out of range of validity.Applying Anderson's argument for a single impurity problem an infinite fixed point is expected. The second order scaling eq. 11 is, however, appropriate to obtain the cross-over temperature T_K as

$$T_K = D\, (v^x\, v^z)^{1/2} \left(\frac{v^x}{4v^z} \right)^{\frac{1}{4v^z}} \qquad 13$$

This result is obtained by integration of eq.11 and it has been assumed that the bare couplings are weak, thus $v^i \ll \frac{1}{4}$. The T_k is very sensitive on the value of v^z if $v^x/v^z \sim 10^{-3}-10^{-4}$. In order to get e.g. $T_K \sim 1K^o$ the v^z must be large enough, thus $v^z \gtrsim 0.3$ for D=10eV. Thus, if $v^z \lesssim 0.2$, then T_K is negligible small, therefore, in this case the commutative model is relevant.

Typical scaling trajectories obtained by numerical integration of eq. 11 are depicted on Fig.s 1 and 2. The change of Δ_o is shown also. It is interesting to note that the many body effects may reduce the value of Δ_o by more than an order of magnitude, thus the distribution function P(E) may be enhanced by the same amplitude.

MEASURABLE QUANTITIES IN THE NON-COMMUTATIVE MODEL

Minimum in the temperature dependence of electrical resistivity: Above T_K the physical quantities can be calculated in the lowest order of perturbation theory, but scaled values of the couplings must be used. Thus, the resistivity is proportional to $(v^x)^2+(v^y)^2+(v^z)^2\big|_{x=T/D}$ which is increasing with lowering the temperature and shows a logarithmic temperature dependence for more than a decade of temperature. In the case of E=0 one can conjecture that at T=0 the electron scattering is determined by the unitarity limit in the two orbital channels $\alpha = 1,2$. The total increase of the resistivity R can be estimated as

$$\Delta R \sim \frac{m}{ne^2} \left(\rho_o^{-1} \frac{2}{\pi} \right) 2\, P_o T_K = \frac{P_o T_K}{N} \frac{1}{e^2} \frac{8\pi}{K_F} \qquad 14$$

where $(\rho_o^{-1} \frac{2}{\pi})$ is the scattering in the unitarity limit, the factor 2 is due to the two channels and $P_o T_K$ is the number of TLS for which $E < T_K$ and therefore the energy splitting does not hinder the formation of resonance (m and e are the electron mass and charge, N is the total number of electrons in unit volume). The expression 14. with $T_K = 5K^o$, $k_F = 1\mathring{A}^{-1}$, $N=10^{-23}/cm^3$ and $P_o=2.10^{18}\ K^{o-1}\ cm^{-3}$ gives $\Delta R \sim 10^{-7}\Omega cm$, which is the order of magnitude observed in many cases (4).

Inelastic electron lifetime: Recently, it has been suggested on the basis of localization theory and of experimental data for thin wires, that in amorphous materials there is an inelastic electron scattering rate τ_{in}^{-1} (16) which is proportional to the temperature T. Black et al.(10) argued that the number of TLS which can be excited by thermal electrons (E<T) is $P_o T$. The golden rule must be applied in the rotated system, in which H_o is diagonal

$$\tau_{in}^{-1} = \frac{4\pi}{h} \{ (\bar{v}^x)^2 + (v^y)^2 \} \rho_o^{-1} P_o T \qquad 15$$

where $(\bar{v}^x)^2 = (v^x)^2 \Delta^2/E^2 + (v^z)^2 \Delta_o^2/E^2$ is the rotated coupling. In order to estimate this expression one can use e.g. $P_o=0.6\ 10^{34} cm^{-3}\ erg^{-1}$ and with a typical medium strong coupling $v^y, \bar{v}^x \sim 0.12$ one gets $\tau_{in}^{-1} = 2.4\ 10^{10} s^{-1} K^{o-1}.T$. The factor T dominates the temperature dependence. The value obtained is of correct order of magnitude suggested by experiments (17). The formation of resonance may resolve the discrepancy quoted earlier (17).

TLS relaxation rates: T_1 and T_2 appearing in the Bloch equation can be measured by ultrasound experiments and they indicate a very fast Korringa relaxation due to the electrons (e.g. 8.9.18), T_1 has been obtained (15) in a similar form to eq. (15) as

$$T_1^{-1} = 16\,\pi\, h^{-1} \{ (v^x)^2 + (\bar{v}^z)^2 \}_{x=T/D} \qquad 16$$

The expression in the curly bracket has been determined for different alloys. If one assumes that v^x, $v^y << v^z$ then one finds $v^z \sim 0.05$ for PdSiCu and for NiP and $v^z \sim 0.16$ for $Pd_{30}Zr_{70}$ (these data are quoted in ref. 9).

CONCLUSION

It has been demonstrated that in metallic glass correlated state can be formed in which the motion of the tunneling atom and the angular dependence of charge oscillation are strongly coupled. The crossover temperature T_K is extremly sensitive on the parameter v^z which can cover a wide range. For PdSiCu alloys T_K is certainly very small and no resistivity minimum due TLS is expected. If v^z is somewhat larger than in $Pd_{30}Zr_{70}$ then the resistivity minimum must occure and the theory predicts a realistic order of magnitude. In this effect only those TLS are effective for which the energy $E \lesssim T_K$. Other interesting effects like the role of TLS in superconductors (see ref. 19) are beyond the scope of the present paper.

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FIGURES

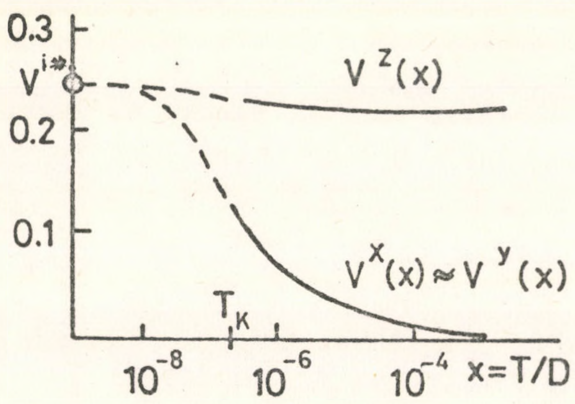

Fig. 1.

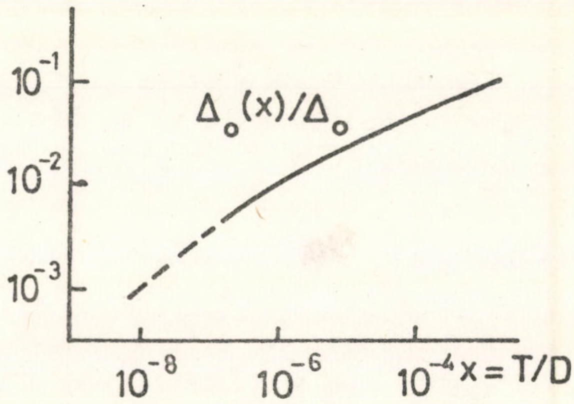

Fig. 2.

Fig. 1. The scaling trajectories calculated numerically using eq. 11 are shown for v^z=0.2 and $v^x/v^z=10^{-3}$. Solid (dotted) curves indicate the parts where the second order scaling is (is not) valid. T_K is calculated from eq. 13. The logarithmic behaviour is appearent around T_K. The fixed point is represented by circle. The region $v^x \sim v^y << v^z$ shows resemblance to ref. 3.

Fig. 2. The change in $\Delta_o(x)$ is depicted with parameters as in Fig. 1.

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