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Experimental tests for the relevance of two-level systems for electron dephasing

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The relevance of tunneling two-level systems (TLS) for electron dephasing in metals is analyzed. We demonstrate that if the concentration of TLS is sufficient to cause the observed dephasing rate, one also should expect quite substantial effects in the specific heat and ultrasound attenuation. In both cases TLS contribution should dominate the electronic one at low enough temperatures.

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Physics of two-level systems (TLS) in disordered metals has been discussed extensively, see for a review Ref. 1 and references therein. Recently this field has been revisited in connection with experiments[2,3] on inelastic relaxation and dephasing of electrons in mesoscopic conductors at low temperatures. Apparent saturation of the inelastic relaxation and dephasing rates at $T\to 0$ is a subject of ongoing discussion. This paper is not a review of the whole spectrum of points of view. We analyze only one of the proposed explanations,[4,5] based on the interaction of conducting electrons with defects possessing internal degree of freedom. Although the specific structure of the defects can be quite complicated, the authors of Refs. 4 and 5 model them by TLS. The TLS-induced dephasing has been related to $1/f$ noise in Ref. 4.

In this paper, we examine this explanation in comparison with the experimental results on the dephasing. In particular, we estimate the TLS concentration, and strength of their coupling with electrons required to describe the experimental data. Both the concentration and the coupling turn out to be large enough to make noticeable effects on other properties of the materials. Namely, we estimate specific heat and ultrasonic attenuation in the presence of TLS with the concentration that follows from the observed dephasing rate. Observation of these effects would be a critical experiment for the TLS-dephasing theory.

Each of the TLS is characterized by a diagonal splitting Δ and tunneling parameter Λ. Introducing Pauli matrices σ_i we can write the TLS Hamiltonian as

$$\tilde{\mathcal{H}}_0=(\Delta/2)\sigma_z+(\Lambda/2)\sigma_x. \tag{1}$$

Here σ_i are the Pauli matrices. It is convenient to change the variables from Δ,Λ to energy splitting between the two levels, $E=\sqrt{\Delta^2+\Lambda^2}$, and projection, $p=(\Lambda/E)^2$. As the very existence of TLS is caused by disorder, the parameters E and p are randomly distributed. Assuming that Δ and $\ln\Lambda$ are uniformly distributed and uncorrelated, we obtain the conventional distribution of E and p for glassy materials, cf., with Ref. 6,

$$\mathcal{P}(E,p)=\frac{P(E)}{2p\sqrt{1-p}}. \tag{2}$$

TLS in glasses are important up to the temperatures $\lesssim 10$ K. In the corresponding energy interval the distribution $P(E)$ is usually assumed to be a constant. Everything we will be speaking about is determined by typical TLS rather than by tails of the distribution. For this reason, it is sufficient to assume that $P(E)$ is constant in a given energy interval and vanishes outside it, $P(E)=(n_T/E^*)\Theta(E^*-E)$. Here n_T has a meaning of the TLS density. In metallic glasses[1] $E^*\gtrsim 10$–20 K, and the TLS density of states n_T/E^* is determined from the experiments on specific heat and acoustic attenuation, $n_T/E^*\approx 10^{16}$ cm^{-3} K^{-1}.

Let us start our discussion with the distribution of these parameters, $\mathcal{P}(E,p)$, which follows from the experimental data on dephasing.[3] The striking result of Ref. 3 is saturation of the dephasing rate τ_φ^{-1} in gold wires as a function of temperature below approximately $T_{\max}=1$ K. The authors claim that $\tau_\varphi(T)$ dependence remains weak down at least to $T_{\min}=40$ mK. Contribution to τ_φ^{-1} of a TLS with $T_{\max}\gtrsim E\gtrsim T_{\min}$ is obviously temperature dependent. Consequently, as it was done in Ref. 4, one has to require the energy distribution to be at least three orders of magnitude narrower than that in metallic glasses, $E^*\lesssim T_{\min}$. It is not impossible that in ordered systems such as crystalline wires TLS are much more similar to each other than in glasses and thus have much narrower parameter distributions. However, the narrowness of the distribution should manifest itself in physical properties.

To describe dephasing by TLS let us include the electron-TLS interaction to Eq. (1). A conventional way[1] is to express it as $\tilde{\mathcal{H}}_{\rm int}=V\sigma_z$, where V has the meaning of the difference between scattering potentials in the ''left'' and ''right'' positions of the defect. After changing the variables to the set (E,p) we have

$$\mathcal{H}_{\rm int}=V\sigma_x\sqrt{p}+V\sigma_z\sqrt{1-p}. \tag{3}$$

As only the first term in Eq. (3) causes dephasing, the contribution of a given TLS is proportional to p. As a result, the dephasing is determined by TLS with $p\sim 1$. This is in contrast with the TLS specific heat where integration over p yields a large logarithmic factor.[6]

In contrast with a static defect, TLS causes inelastic scattering of electrons, the energy transfer being E. The inelastic relaxation rate $1/\tau_{\rm in}$ can be expressed through the inelastic scattering cross section, $\sigma_{\rm in}\equiv 4\pi\chi/k_F^2$ as

$$1/\tau_{\rm in}=v_F\sigma_{\rm in}n_T=4\pi\chi v_F n_T/k_F^2. \tag{4}$$

Here v_F and $\hbar k_F$ are the Fermi velocity and momentum, respectively. Important dimensionless parameter $\chi\leqslant 1$ has the meaning of the inelastic cross section in units of its unitary limit $4\pi/k_F^2$. It follows from Eq. (4) that the ratio between the TLS concentration n_T and the electron concentration $n_e=k_F^3/3\pi^2$ can be estimated as

$$n_T/n_e=(3\pi/8\chi)(\hbar/\epsilon_F\tau_{\rm in}). \tag{5}$$

Provided that $E^*\tau_\varphi\gg\hbar$, each inelastic scattering event causes dephasing. Therefore, the dephasing rate $\tau_\varphi\approx\tau_{\rm in}$. In the opposite limit, $E^*\tau_\varphi\ll\hbar$, many inelastic scattering events are needed to destroy phase coherence. As a result, in general case[7,8]

$$\frac{\tau_{\rm in}}{\tau_\varphi}\sim\begin{cases}1, & E^*\tau_{\rm in}\gg\hbar\\ (E^*\tau_{\rm in}/\hbar)^{2/3}, & E^*\tau_{\rm in}\ll\hbar.\end{cases} \tag{6}$$

Assuming $\tau_\varphi=3$ ns and $E^*\approx 20$ mK$>\hbar/(3,$ ns$)$, we obtain for $\epsilon_F=5$ eV the estimate $n_T/n_e\approx 3\times 10^{-8}$. For $n_e\approx 3\times 10^{22}$ cm^{-3} the TLS density of states turns out to be of the order of $(n_T/E^*,\text{cm}^{-3}\ \text{K}^{-1})\approx 5\times 10^{16}/\chi$. At $E^*\tau_\varphi\ll\hbar$ the above estimate should be multiplied by a large factor $(\hbar/E^*\tau_\varphi)^2\gg 1$.

Let us compare the contributions to the specific heat of TLS, C_T, and electrons, C_e:

$$C_e=3n_eT/2\epsilon_F, \tag{7}$$

$$C_T=\frac{n_T T\mathcal{L}}{E^*}f\left(\frac{E^*}{T}\right),\quad f(x)=\begin{cases}1, & x\ll 1\\ x^3/12, & x\ll 1.\end{cases} \tag{8}$$

Large factor $\mathcal{L}\gg 1$ appears in Eq. (8) due to the fact that, in contrast with the case of $1/\tau_\varphi$, even very ''slow'' TLS contribute to the specific heat. We think that this fact was not fully appreciated in Refs. 4 and 5. Formally, $\mathcal{L}$ originates from the logarithmic divergence of the integral $\int\mathcal{P}(E,p)dp$ at $p\to 0$. The actual limits of this integration are not well understood. Usually this factor is estimated as $\mathcal{L}=\ln(t_{\rm exp}/\tau_{\rm min})$, where $t_{\rm exp}$ is the measurement time while $\tau_{\rm min}$ in the minimal relaxation time of TLS population. A realistic estimate is $\mathcal{L}\approx 20$–40.

It follows from Eqs. (7), (8), and (5) that at high temperatures C_T is negligible, whereas at $T\ll E^*$

$$C_T/C_e\approx(\mathcal{L}/\chi)(\hbar/E^*\tau_{\rm in}). \tag{9}$$

For our example, $C_T/C_e\approx 0.1\mathcal{L}/\chi$.

The dimensionless cross section χ can be estimated from relaxation acoustic attenuation, see, e.g., Ref. 1 and discussion below. In metallic glasses such an estimation gives $\chi=0.01$–0.1. Taking $\mathcal{L}=30$, $\chi=0.03$ we conclude that C_T at low enough temperatures exceeds C_e by *about a hundred times*. At the same time, as $T\gg E^*$ the TLS contribution rapidly decreases with temperature increase. Consequently, the ratio C/T changes dramatically at $T\approx E^*$, being almost constant both at $T\ll E^*$ and $T\gg E^*$. This effect provides a possibility to experimentally determine both E^* and n_T.

Another independent way to determine parameters of TLS is the study of the acoustic attenuation. The advantage is that parameters χ and E^* are relevant even at $T\gg E^*$. Moreover, it is well known[1] that there is almost no ultrasound attenuation due to free electrons and phonons at low enough acoustic frequency and temperature. Therefore even low concentration of TLS provides dominating contribution.

There are two contributions of TLS to the attenuation.[9] The first one is a direct interlevel absorption of acoustic quanta known as the resonant mechanism. The second, relaxation mechanism arises from acoustic wave-induced time dependence of the energy splitting E. Nonequilibrium component of the TLS population, which is caused by this dependence, relaxes due to electron-TLS interaction. This relaxation produces acoustic attenuation.

To evaluate the power W dissipated per unit volume of the metal we make a usual assumption[9] that the ultrasound affects only the diagonal part of the Hamiltonian Eq. (1): $\Delta\sigma_z$ is substituted by $(\Delta+a\cos\omega t)\sigma_z$ where ω is the ultrasound frequency. In the basis where the unperturbed Hamiltonian Eq. (1) is diagonal, the first-order perturbation can be written as a sum $a\cos\omega t(\sigma_z\sqrt{1-p}+\sigma_x\sqrt{p})$. The two terms lead, respectively, to the relaxation and resonant attenuation, $W_{\rm rel}$ and $W_{\rm res}$. Note that the off-diagonal part of the perturbation vanishes at $p\to 0$, and thus the integral over p is determined by $p\sim 1$. Therefore $W_{\rm res}$ in contrast with C_T does not contain the large logarithmic factor $\mathcal{L}$. This factor does not appear in $W_{\rm rel}$ either for the same reason as it did not appear in $1/\tau_\varphi$–TLS-electron relaxation rates vanish at $p\to 0$.

In the absence of TLS relaxation one can calculate the contribution to W from one impurity i characterized by (p_i,E_i) using the Fermi golden rule

$$\delta W_{\rm res}^{(i)}(\omega)=(\pi a^2/2)\omega p_i\tanh(E_i/2T)\,\delta(\hbar\omega-E_i). \tag{10}$$

Here $\hbar\omega$ is the energy transfer, and the factor p_i appears because the transition matrix element is proportional to the tunneling coupling Λ, while the factor $\tanh(E_i/2T)$ is the occupation difference of the two levels with the distance $E_i=\hbar\omega$.

Interaction of TLS with electrons broadens the resonance. One can take this broadening into account replacing the δ function in Eq. (10) by Lorentzian $\pi^{-1}\Gamma^{(i)}/[(\hbar\omega-E^{(i)})^2+(\Gamma^{(i)})^2]$. Here Γ is the TLS off-diagonal relaxation rate which is an analog of the rate $\hbar/T_2$ in the physics of spin resonances. As each inelastic process leads to simultaneous relaxation of a TLS and an electron, the rate Γ is directly related to the electron inelastic relaxation rate, Eq. (4), and thus is determined by the same dimensionless inelastic cross section χ, see Ref. 1,

$$\Gamma^{(i)}=\chi p_i E_i/[2\tanh(E_i/2T)]. \tag{11}$$

The relaxation contribution is given by the Debye-type formula, see, e.g., Ref. 10,

$$\delta W_{\rm rel}^{(i)} = \frac{\omega a^2}{2} \frac{\omega(1-p)}{4T\cosh^2(E_i/2T)} \cdot \frac{2\Gamma^{(i)}/\hbar}{\omega^2 + (2\Gamma^{(i)}/\hbar)^2}. \quad (12)$$

The total dissipated power per unit volume is obtained by the averaging of Eqs. (10) and (12) over p_i and E_i with the distribution (2). For presenting the results, we introduce dimensionless frequency $\Omega = \hbar\omega/E^*$ and temperature $\theta 2T/E^*$. In these terms the total dissipated power per unit volume W has the form

$$W = (n_T a^2/2\hbar)\Omega F(\chi,\Omega,\theta), \quad F = F_{\rm res} + F_{\rm rel}, \quad (13)$$

where the dimensionless functions are given by

$$F_{\rm res} = \int_0^1 \int_0^1 \frac{dx dp \chi x p (1-p)^{-1/2}}{4(x-\Omega)^2 + (\chi x p)^2 \tanh^{-2}(x/\theta)}, \quad (14)$$

$$F_{\rm rel} = \int_0^{1/\theta} \frac{\chi\theta y dy}{2\cosh^3 y} \int_0^1 \frac{dp\sqrt{1-p}\Omega \sinh y}{\Omega^2 \tanh^2 y + (\chi p y \theta)^2}. \quad (15)$$

Equations (13)–(15) determine frequency and temperature dependences of the ultrasound attenuation. We see that W depends upon two tunable parameters, Ω and θ. Thus the investigation of frequency and temperature dependences of $Q_{\rm res}$ provides a possibility to extract E^* and χ, see below Eqs. (16) and (17), without an independent measurement of parameter a.

First, we analyze the relaxation attenuation. Since in Eq. (15) only $y \lesssim 1$ and $p < 1$ are important, to make an estimate one can expand $\tanh y$ and neglect p in $\sqrt{1-p}$. The result can be conveniently approximated as

$$F_{\rm rel} \approx (\chi/\tilde{\Omega})\min\{\theta,1\}, \quad \tilde{\Omega} = \max\{\Omega, \chi\theta\}. \quad (16)$$

Similar approximation of Eq. (14) yields

$$F_{\rm res} = \begin{cases} -\chi \ln\tilde{\Omega} + \Omega/\max\{\Omega,\theta\}, & \tilde{\Omega} \ll 1 \\ \chi/\tilde{\Omega}^2, & \tilde{\Omega} \gg 1. \end{cases} \quad (17)$$

As one can see, at high frequencies and temperatures the relaxation mechanism dominates, whereas at lowest temperatures the resonant mechanism is more important. It should be noted that the two contributions to W can be effectively separated in experimental studies of nonlinear attenuation. Indeed, $W_{\rm res}$ is well known,[9,1] to be suppressed by relatively weak ultrasound, so that only the relaxation contribution remains. According to Eqs. (16), the frequency and temperature dependences of the attenuation differ qualitatively for $\hbar\omega \gtrsim \chi T$ and $\hbar\omega \lesssim \chi T$ and for $T < E^*$ and $T > E^*$. This should enable us to determine the parameters χ and E^*.

Let us compare the estimated value of W with the acoustic attenuation in conventional metallic glasses W_g assuming that the parameters a and χ are the same. It turns out that the resonant contributions are related as TLS densities of states at the energy $\hbar\omega$. We have already concluded that the explanation[4] of the experiments[3] requires the TLS density of states n_T/E^* to exceed its typical value for metallic glasses P_g by *one–two orders of magnitude*. As in metallic glasses the acoustic attenuation has been successfully measured, one should expect rather large effect. Comparing the relaxation contributions one finds $W_{\rm rel}/W_g \approx (n_T/P_g \max\{E^*,T\}) \simeq 10 \div 10^2 \min(1, E^*/T)$.

According to several publications, see Refs. 11 and 12 for a review, in real metals interaction of TLS with electrons may cause two-channel Kondo (2CK) effect with rather high (1–10 K) Kondo temperature, T_K. For $T < T_K$, the cross section for inelastic scattering reaches the unitary limit, $\chi = 1$ in Eq. (4). It was suggested in Ref. 5 that the experimentally observed temperature independent dephasing rate is a manifestation of the 2CK effect. We are convinced that possible values of T_K in real metals are several orders of magnitude lower than the estimates of Refs. 13 and we presented a detailed discussion in a separate paper.[14] We believe that electron-hole asymmetry discussed by Zawadowski and Zárand[13] in cond-mat/009283 and does not change the above conclusion, see detailed discussion in our cond-mat/0102513. Nevertheless, in a context of a phenomenological speculation it makes sense to examine the assumptions behind the explanation of Ref. 5 and to discuss whether those assumptions manifest themselves in other observable quantities such as the specific heat and sound attenuation.

Let us adopt the simplistic model of Ref. 5 and characterize the TLS's by randomly distributed Kondo temperatures T_K and the ''bare'' level splitting Δ. The distribution $\mathcal{P}(T_K,\Delta)$ can be written as

$$\mathcal{P}(T_K,\Delta) = (n_T/\Delta^* T_K^*) P(T_K/T_K^*, \Delta/\Delta^*), \quad (18)$$

where T_K^* and Δ^* are some cutoff values for the Kondo temperature and the splitting, respectively. The dimensionless function $P(x,y)$ is assumed to decay rapidly enough at $x \gg 1$ or $y \gg 1$ and to be normalized, $\int_0^\infty dx \int_0^\infty dy P(x,y) = 1$. In particular, we neglect the effects of the anisotropy in the coupling constants of TLS's with electrons. This simplification does not change final results qualitatively.

Each TLS is characterized by its inelastic cross section,

$$\sigma_{\rm in}^{(i)} = (4\pi/k_F^2)\tilde{\chi}(T_K^{(i)}/T, \Delta^{(i)}/T), \quad (19)$$

where superscript (i) labels a particular TLS, and $\tilde{\chi}(x,y)$ is a dimensionless function with the following asymptotic behavior (we omitted numerical prefactors):

$$\tilde{\chi}(x,y) = \begin{cases} 1, & x \gg 1, \quad y^2 \ll x; \\ x^2/y^4, & x \gg 1, \quad x \ll y^2 \ll x^2; \\ L, & \text{otherwise}; \end{cases} \quad (20)$$

$$L \equiv [\ln\max\{1/x, y/x\}\max(1, y\ln\max\{1/x, y/x\})]^{-2}.$$

Combining Eqs. (19) and (18), one obtains for the dephasing time τ_φ the expression

$$\frac{\tau_0}{\tau_\varphi} = \int dx dy P(x,y) \tilde{\chi}\left(x\frac{T_K^*}{T}, y\frac{\Delta^*}{T}\right), \quad (21)$$

where $1/\tau_0 = 4\pi n_T v_F/k_F^2$ is the maximal rate of the electron scattering off a TLS. Using asymptotic expressions (20) in Eq. (21) we find

$$\frac{\tau_0}{\tau_\varphi}=\begin{cases}\dfrac{\min(1,T/\Delta^*)}{\ln^2[\max(T/T_K^*,\Delta^*/T_K^*)]}, & T\gg T_K^*;\\ \min\{1,(TT_K^*)^{1/2}/\Delta^*\}, & T\ll T_K^*.\end{cases}\tag{22}$$

According to Eq. (22), the saturation in temperature dependence of τ_φ is possible *only* if $\Delta^*<T_K^*$. The corresponding temperature region is $(\Delta^*/T_K^*)^2<T/T_K^*<1$. Temperature saturation of τ_φ was observed in Ref. 3 in the interval 40 mK$<T<$1 K. This indicates that the distribution of the level splittings required for the phenomenology of Ref. 5 is *not typical for metallic glasses* but rather narrow with the upper boundary $\Delta^*\simeq 200$ mK. Therefore, the assumption about unusually narrow level splitting distribution is intrinsic both for Refs. 4 and 5. In this respect, the difference in physical assumptions in these references appears quantitative rather than qualitative—both interpretations of the data[3] imply that TLS's in the gold wires are essentially different from those in metallic glasses.

Accepting the model,[5] let us now discuss the specific heat and acoustic absorption. The specific heat for a particular TLS $C^{(i)}\simeq(T/T_K^{(i)})\ln T_K^{(i)}/T$ at $(\Delta^{(i)})^2/T_K^{(i)}\ll T\ll T_K^{(i)}$.[15] It yields the specific heat similar to Eq. (9) with $\chi=\mathcal{L}=1$ and $E^*\simeq T_K^*$. Therefore, the TLS can produce the correction to the specific heat of the order of 2%, which is difficult to detect.

Sound attenuation of the TLS in Kondo regime happens mostly due to the relaxation mechanism. The relaxation rate (11) for the individual impurity (i) in the regime $\min\{\Delta^{(i)},(\Delta^{(i)})^2/T_K^{(i)}\}\ll T$ can be estimated as $\Gamma^{(i)}=\hbar^{-1}\max\{T/\ln^2(T/T_K^{(i)}),T_K^{(i)}\}$. Using such rate and $p=0$ in Eq. (12), and averaging the result with the help of Eq. (18), we obtain for the most realistic range of frequencies $\hbar\omega\ll T$

$$W\approx\frac{n_T a^2\omega}{T}\frac{\hbar\omega}{T_K^*}\begin{cases}1, & T<T_K^*;\\ \ln^{-2}T/T_K^*, & T>T_K^*.\end{cases}\tag{23}$$

This value is smaller than the corresponding result in glasses by a factor $\hbar\omega/T_K^*$, which once again makes it difficult to observe.

In conclusion, we demonstrated that if the concentration of TLS is sufficient to cause the observed dephasing rate, one also should expect quite substantial and specific effects in the specific heat C and ultrasound attenuation W. In both cases TLS contribution should dominate the electronic one at low enough temperatures. These effects persist provided that $T\gg T_K^*$, where T_K^* is the mean Kondo temperature. In the opposite limit, which corresponds to the developed two-channel Kondo effect, TLS effects on C and W seem to be relatively small. We do not think, however, that the limit $T\ll\overline{T}_K^*$ is possible to realize at the experimentally accessible temperatures, see Ref. 14 for the detailed discussion. For this reason we believe that the absence of the TLS contributions to C and W would mean their irrelevance for the dephasing rather than a realization of the two-channel Kondo scenario.

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