

TEMPERATURE DEPENDENCE OF THE EPR LINE WIDTH FOR CONDUCTION ELECTRONS
IN METALS CONTAINING NONMAGNETIC IMPURITIES

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The spin-lattice relaxation time T_1 of the conduction electrons in metals with impurities is calculated with allowance for the thermal vibrations of the impurities. It is shown that at low temperatures the impurity time of the relaxation times (for heavy impurities) is practically independent of the temperature, and at high temperatures it varies like $a - b/T$ ($a, b > 0$). Expressions are presented for the temperature dependence of the contributions made to T_1 by the scattering of the electrons by the deformed phonon spectrum of the lattice and by the interference between the scattering from the vibrating impurity and from the vibrations of the host lattice, for different relations between the masses of the impurity and the host atom and their amplitude of inelastic spin scattering.

RESONANT paramagnetic absorption by conduction electrons is observed in a relatively small number of metals (Li, K, Rb, Cs, Be)^[1-4]. It is assumed that in metals the main relaxation mechanism, which determines the width of the EPR line for the conduction electrons, is modulation of the spin-orbit interaction V by the thermal vibrations. For valence electrons, V increases rapidly with increasing atomic number Z , although no simple expression describing the $V(z)$ dependence has been described as yet. For free lithium ($Z = 3$), for example, the spin orbit splitting of the valence level $2p$ is 2.77×10^{-5} eV, whereas for sodium ($Z = 11$) its value is 2.12×10^{-3} eV^[4]. From the measurements of the impurity contribution to the line width $\Delta H'$ of the paramagnetic resonance on conduction electrons (PRCE) in alkali metals^[5-7] it follows that the spin-orbit coupling of the conduction electron with the impurity atom in the metal does not differ in order of magnitude from the values for the free atom.

In investigations of the PRCE line width, the impurity atoms are usually regarded as static defects^[6,8,9]. This causes the width $\Delta H'$ to be proportional to the concentration of the impurities and to be independent of the temperature—the analog of the Mathiessen rule in the theory of electric conductivity. Allowance for the vibrations of impurity atoms, and in some cases the appreciable differences between the thermal vibrations of a crystal with impurities and the vibration spectrum of an ideal crystal, have made it possible to explain in a number of recent papers^[10-13] some of the experimental results (violation of the Mathiessen rule in weak solutions, the anomalies of the lattice specific heat).

Obviously, allowance for the impurity vibrations in the calculation of the impurity contribution to the PRCE line width should lead to the appearance of a temperature dependence of $\Delta H'$. Since the magnitude of the spin-orbit interaction changes strongly with changing Z , the contribution of the thermal vibrations to $\Delta H'$ can be expected to be pronounced quantitatively more than in measurements of the temperature dependence of the electric resistance.

We present in this paper a calculation of the spin-

lattice relaxation time of the conduction electrons in a metal with nonmagnetic impurities. The spin-lattice relaxation time T_1 , connected with the peak width of the symmetrical PRCE line by the relation^[14]

$$\Delta H = 2\hbar / \sqrt{3} T_1 \mu_e, \tag{1}$$

is calculated by means of the formula

$$\frac{1}{T_1} = \frac{4}{(2\pi)^3} \left(\int \frac{\partial f_\beta(k)}{\partial E_k} dk \right)^{-1} \int \frac{\partial f_\beta(k)}{\partial E_k} \times [(1 - f_\alpha(k')) W_{k'\alpha, k\beta} + f_\alpha(k') W_{k\beta, k'\alpha}] dk dk', \tag{2}$$

where $W_{k'\alpha, k\beta}$ is the probability of the transition of the electron upon scattering from the state with wave vector k and spin β into the state $k'\alpha$; $f(k)$ is the electron distribution function.

In the calculation of $W_{k'\alpha, k\beta}$ in the Born approximation we use the well known relation^[15]

$$W_{k'h} = S(q, \omega) / \hbar^2. \tag{3}$$

Here $S(q, \omega)$ is the scattering correlation function

$$S(q, \omega) = \int dt e^{i\omega t} \langle V_{j\gamma}^*(q, t) V_{i\eta}(q) \rangle, \tag{4}$$

$\omega = \omega_{k\beta} - \omega_{k'\alpha}$, $V_j(q)$ is the Fourier component of the operator of spin-orbit interaction between the electron and the ion of species γ in the j -th site of the lattice, and the symbol $\langle \dots \rangle$ denotes averaging over the Gibbs ensemble. Since the ions of different source are located in the lattice sites randomly, we assume that averaging over the ensemble includes averaging over all possible ion configurations. For a crystal containing impurities of one species (c —impurity concentration), when the internal state of the ion does not change during the collision,

$$V_{j\gamma}(q, t) = V_{1,0}(q) e^{i\mathbf{q}\mathbf{R}_j(t)}, \quad \mathbf{R}_j(t) = \mathbf{R}_j(0) + \mathbf{u}_j(t), \tag{5}$$

$\mathbf{u}_j(t)$ —operator of displacement of the ion from the equilibrium position, $\gamma = 1$ or 0 . Confining ourselves to single-phonon scattering processes we obtain for $W_{k'\alpha, k\beta}$

$$W_{k'h} = W(1) + W(2) + W(3), \tag{6}$$

$$W_{k'h}(1) = \hbar^{-2} N c |V_1(q)|^2 [2\pi\delta(\omega^2) + \langle (\mathbf{q}\mathbf{u}_1(t)) (\mathbf{q}\mathbf{u}_1(0)) \rangle_\omega],$$

$$W_{kh}(2) = \hbar^{-2} N c |V_1(q) V_0(q)| \left\langle \sum_j \exp(iqR_j) (\mathbf{q}u_i(t)) (\mathbf{q}u_0(0)) \right\rangle_\omega, \quad (7)$$

$$W_{kh}(3) = \hbar^{-2} N c |V_0(q)|^2 \left\langle \sum_{i,j} \exp(iq(\mathbf{R}_i - \mathbf{R}_j)) (\mathbf{q}u_{i0}(t)) (\mathbf{q}u_{j0}(0)) \right\rangle_\omega \quad (8)$$

Expressions (6)–(8) describe respectively the impurity scattering, interference scattering, and scattering by the deformed phonon spectrum of the lattice.

The Fourier components of the correlation functions in (6)–(8) can be expressed in terms of the Fourier components of the Green's function

$$G_{ij}^{\alpha\beta}(t) = -i\theta(t) \langle u_i^\alpha(t) u_j^\beta(0) - u_j^\beta(0) u_i^\alpha(t) \rangle, \quad \alpha, \beta = x, y, z \quad (9)$$

with the aid of the relation^[15]:

$$\langle u_i^\alpha(t) u_j^\beta(0) \rangle = -2(1 - e^{-p\omega})^{-1} \text{Im} G_{ij}^{\alpha\beta}(\omega + i\delta), \quad p = \hbar/kT. \quad (10)$$

The equations for the Green's functions (9) for crystals with impurities that differ in mass from the host atoms (impurity of the isotopic type) were considered in^[12, 16-18]. It is convenient here to use the expressions given by Kagan and Zhernov^[11] for the sought Fourier components of the correlation functions. For an isotropic crystal, they take on respectively the values:

$$\frac{2\pi\hbar}{M} q^2 \frac{\text{sign } \omega}{1 - e^{-p\omega}} \varphi(\omega^2), \quad (6')$$

$$\frac{2\pi\hbar}{M} q^2 \frac{\text{sign } \omega}{1 - e^{-p\omega}} \left[\frac{1}{\pi} \frac{P}{\omega^2 - \omega_q^2} \gamma(\omega^2) + \delta(\omega^2 - \omega_q^2) \right], \quad (7')$$

$$\frac{2\pi\hbar}{M} q^2 \frac{\text{sign } \omega}{1 - e^{-p\omega}} \frac{1}{\pi} \frac{c\epsilon\omega^2\gamma(\omega^2)}{[\omega^2(1 - c\epsilon\Delta(\omega^2)) - \omega_q^2]^2 + [c\epsilon\omega^2\gamma(\omega^2)]^2}, \quad (8')$$

where $\epsilon = (M - M')/M$, M' is the mass of the impurity atom, M is the mass of the atom of the ideal lattice, and P is the symbol of the principal value;

$$R(\omega^2) = (1 - \epsilon\omega^2 J(\omega^2))^2 + (\pi\epsilon\omega^2 g(\omega^2))^2, \quad (11)$$

$$J(\omega^2) = P \int_0^{\omega^2} dz \frac{g(z)}{z - \omega^2}, \quad \varphi(\omega^2) = \frac{g(\omega^2)}{R(\omega^2)},$$

$g(\omega^2)$ is the distribution function of the squares of the phonon-spectrum frequencies of the ideal lattice, and

$$\gamma(\omega^2) = \frac{\pi\epsilon\omega^2 g(\omega^2)}{R(\omega^2)}, \quad \Delta(\omega^2) = \frac{1 - \epsilon\omega^2 J(\omega^2)}{R(\omega^2)}.$$

Substituting the expression for the transition probabilities with allowance for relations (6')–(8') into the formula for T_1^{-1} , we get

$$\frac{1}{T_1} = \frac{1}{T_1(1)} + \frac{1}{T_1(2)} + \frac{1}{T_1(3)}, \quad (12)$$

where

$$\frac{1}{T_1(1)} = cr \left(\int_0^{q_0} b_1(q) q dq + \frac{4\hbar}{M} \int_0^{q_0} b_1(q) q^3 dq \int_0^\infty \frac{e^{p\omega}}{e^{2p\omega} - 1} \varphi(\omega^2) d\omega \right), \quad (13)$$

$$\frac{1}{T_1(2)} = cr \frac{4\hbar}{M} \int_0^{q_0} a(q) q^3 dq \int_0^\infty \frac{e^{p\omega}}{e^{2p\omega} - 1} \times \left[\frac{\pi^{-1}P}{\omega^2 - \omega_q^2} \gamma(\omega^2) + \Delta(\omega^2) \delta(\omega^2 - \omega_q^2) \right] d\omega, \quad (14)$$

$$\frac{1}{T_1(3)} = r \frac{4\hbar}{M} \int_0^{q_0} b_0(q) q^3 dq \times \int_0^\infty \frac{e^{p\omega}}{e^{2p\omega} - 1} \frac{\pi^{-1}c\epsilon\omega^2\gamma(\omega^2) d\omega}{[\omega^2(1 - c\epsilon\Delta(\omega^2)) - \omega_q^2]^2 + [c\epsilon\omega^2\gamma(\omega^2)]^2}, \quad (15)$$

$r = 4\pi\hbar^{-1}\rho(E_F)N$, $p = \hbar/kT$, N —number of particles in the sample, $b_{1,0} = |V_{1,0}(q)|^2$, $\rho(E_F)$ —density of electrons near the Fermi level, and $a(q) = |V_1(q)V_0(q)|$. In the derivation of these equations we used the customary assumptions in concerning the phonon spectrum of an ideal crystal: $\omega = qs$, q —wave vector of the phonon, s —speed of sound in the sample, $q_0 = q_{\text{max}} = 2k_F$, k_F —momentum of electron with Fermi energy E_F , and $\omega_0 = q_0s$.

If we include in the elastic part of the impurity scattering in (13) the factor $\exp(-W_1)$, which depends on the Debye-Waller factor

$$W_1(q) = q^2 \frac{\hbar}{M} \int_0^\infty (2n(\omega) + 1) \varphi(\omega^2) d\omega,$$

then, when $W_1 < 1$, we obtain for the temperature-dependent part of the impurity scattering

$$\frac{1}{T_1(1)} = -rc \frac{\hbar}{M} \int b_1(q) q^3 dq \int_0^\infty \text{th} \frac{p\omega}{2} \varphi(\omega^2) d\omega. \quad (13')$$

We shall find it useful to have a table of the numerical values of the parameters contained in the relations for T_1^{-1} . In the table, Δ is the spin-orbit splitting of the valence electron of the free atom^[4] and A is the atomic weight. According to estimates given in^[5, 6, 8], $V \approx \Delta$ for impurities.

	A	Δ , eV
Li	6.9	$2.77 \cdot 10^{-5}$
Be	9	$3.72 \cdot 10^{-4}$
Na	23	$2.12 \cdot 10^{-5}$
Zn	65	$7.17 \cdot 10^{-2}$
Cd	112	0.212
Tl	204	0.96

1. Let us consider the case of a heavy impurity, when $\epsilon < 0$. This was precisely the case realized in all the listed experimental papers on the influence of impurities on the width of the PRCE lines^[5-7]. Unfortunately, the measurements of $\Delta H'$ were made in these investigations at a fixed temperature ($T \approx 293^\circ\text{K}$). When $|\epsilon| > 1$ in (13') the function $\varphi(\omega^2)$, which plays the role of the distribution function of the squares of the frequencies in the impurity vibration spectrum, has a resonant character with a maximum in the region of the quasi local frequency, equal to

$$\omega_* = \left[|\epsilon| \int \frac{g(\omega^2)}{\omega^2} d\omega \right]^{-1/2}.$$

In the region of low temperatures $T < \hbar\omega_*/k$, $g(\omega^2) \approx 3\omega/2\omega_0^2$,

$$\frac{1}{T_1(1)} = -cr \frac{9|\epsilon|}{2\pi^2\omega_0} \frac{\hbar}{M} \int b_1(q) q^3 dq,$$

$$\frac{1}{T_1(2)} = cr \frac{\hbar}{M} \int a(q) q^3 dq \left\{ \frac{2}{\omega_q} \frac{e^{p\omega_q}}{e^{2p\omega_q} - 1} - \frac{6\epsilon}{\omega_q^2} \left(\frac{T}{\Theta} \right)^3 \frac{1}{p} J_4 \right\}. \quad (16)$$

For small impurity concentrations, the function that depends on c in the integrand of (15) can be represented in the form

$$\Phi(\omega, c) \approx \delta(\omega^2 - \omega_q^2) - 2c\epsilon\omega\delta'(\omega^2 - \omega_q^2) + \frac{1}{\pi} c\epsilon\omega^2\gamma(\omega^2) \frac{P}{\omega^2 - \omega_q^2} \quad (17)$$

It is easy to see that the integration with respect to ω in the relation for T_1^{-1} (3), with allowance for the first

term in (17), yields an expression for the spin-lattice relaxation due to the vibrations of the crystal without the impurity. The contribution made to $\Delta H'$ as a result of the distortion of the character of the vibration of the ideal crystal by the impurities is described by the remaining two terms in (17). Retaining only the principal terms in the integration with respect to ω in (15), we obtain the impurity part in $1/T_1(3)$

$$(-2c\epsilon)r \int b_0(q)q^3 dq \left[\frac{e^{p\omega_q}}{e^{2p\omega_q} - 1} \left(\frac{1}{\omega_q} - p \right) - \frac{3\epsilon}{4} \frac{1}{\omega_q^4 p^3} \left(\frac{T}{\Theta} \right)^3 J_6 \right], \quad (18)$$

where $J_n = \Gamma(n)\zeta(n)$, $\zeta(n)$ is the Riemann zeta function, $\Gamma(n) = (n-1)!$, and Θ is the Debye temperature.

Unfortunately, there are no investigations from which it is possible to draw any conclusions concerning the dependence of $V(q)$ on q . We shall therefore assume that

$$V_{1,0}(q) = A_{1,0}, \quad A_V = \Delta_V.$$

Qualitative results obtained under this assumption remain unchanged if

$$V(q) = \sum a_n q^n, \quad n \geq 0$$

and $V(q) > 0$ for all values of q . Calculating (16) and (18), we get

$$\frac{1}{T_1(1)} = -cr \frac{9|\epsilon|}{8\pi^2} A_1^2 \frac{q^4}{\omega_0} \frac{\hbar}{M}, \quad (19)$$

$$\frac{1}{T_1(2)} = cr A_1 A_0 \frac{q_0^4}{\omega_0} \frac{\hbar}{M} \left(\frac{T}{\Theta} \right)^3 J_3 \left[2 - 3\epsilon \left(\frac{T}{\Theta} \right) \frac{J_4}{J_3} \right], \quad (20)$$

$$\frac{1}{T_1(3)} = 2cer A_0^2 \frac{q_0^4}{\omega_0} \frac{\hbar}{M} \left(\frac{T}{\Theta} \right)^3 J_3 \left[1 - \frac{J_4}{J_3} - \frac{3\epsilon}{4} \left(\frac{T}{\Theta} \right) \ln q_0 \frac{J_6}{J_3} \right]. \quad (21)$$

As seen from the table, for heavy impurities forming true solid solutions with alkali metals (the alkali metals do not form such solutions with one another in the solid state^[20]) we have $A_1 \gg A_0$. Therefore in this case the main contribution to $\Delta H'$ is made by the quantity $1/T_1(1)$. In the region $T \ll \Theta$, the contribution to $\Delta H'$ due to the impurity vibrations ceases to depend on the temperature, as follows from (15) and (19). Physically this result can be attributed to the fact that the decrease of the probability of elastic scattering with increasing temperature, due to the Debye-Waller factor, is fully compensated by the increase in the probability of inelastic scattering. As a net result, the contribution to $\Delta H'$ is made by the low-frequency part of the zero-point oscillations, the amplitude of which for the impurity increases by a factor

$$\varphi(\omega^2) / g(\omega^2)$$

compared with the amplitude of the low-frequency vibrations of the atoms of the ideal crystal.

The relaxation times due to the scattering by the immobile impurity, $T_1(0)$ and by the ideal-lattice vibrations have in the assumed approximation the following values:

$$\frac{1}{T_1(0)} = cr A_1^2 \frac{q_0^2}{2}, \quad (22)$$

$$\frac{1}{T_{10}} = r A_0^2 \frac{q_0^4}{\omega_0} \frac{\hbar}{M} \left(\frac{T}{\Theta} \right)^3 J_3.$$

For samples of lithium containing Zn or Cd impurities

with concentration $c \approx 2-5$ at.%, used in the measurement of the PRCE line width in^[5], it is easy to see that in the temperature interval $0 \leq T \leq T_{\text{melt}}$ (T_{melt} — melting temperature) we have

$$T_1(0) / T_{10} \leq 10^{-3}.$$

Substituting in (16) and (22) the values of the parameters listed in the table ($q_0 = 3.2 \times 10^8$ cm⁻¹ for Li and 10^8 cm⁻¹ for Na) for the samples of Li + Cd and Na + Tl, we get $\alpha = T_1(0)/T_1(1)$, $\alpha(\text{Li}) = -0.83$, and $\alpha(\text{Na}) = -0.12$. Thus, at low temperatures, the contribution made to the impurity width of the PRCE lines by the part that depends on the impurity vibrations is of the same order as the width due to the scattering by the immobile impurity.

The ratio $A_1/A_2 \leq 10$ is realized, as can be seen from the table, under the condition $|\epsilon| \leq 1$ (the impurities of Be and Li). In this case the quantity $T_1^{-1} - T_{10}^{-1}$ will increase with temperature like $T_1^{-1}(2)$, and the temperature dependent part will be approximately 1/10 of $T_1^{-1}(0)$.

Finally, let us analyze the temperature dependence of that part of $\Delta H'$ which is due to the deformation of the lattice vibration spectrum by the impurities (21). The most convenient samples for the separation of this part against the total PRCE line width are alloys of metals with close values of the spin-orbit coupling constants, since the $T_1^{-1}(1)$ and $T_1^{-1}(3)$ contributions depend on the concentration and are proportional to A_1^2 and A_0^2 , respectively.

When $T/\Theta < 0.1$, the term containing J_5 in (21) can be neglected. The remaining expression

$$\frac{1}{T_1(3)} = \frac{1}{T_{10}} (-2c\epsilon) \left(\frac{J_4}{J_3} - 1 \right) \quad (21')$$

coincides qualitatively with the analogous expression for the resistance in the theory of the electric conductivity of metals with nonmagnetic impurities, developed by Kagan and Zhernov^[11]. It can thus be assumed that the deformation of the lattice spectrum by heavy impurities leads to an increase in the density of the number of phonons in the low-frequency region.

With increasing temperature, $T_1^{-1}(3)$ increases monotonically and reaches a maximum value at a temperature

$$T_m = \Theta \sqrt{\frac{1}{15|\epsilon|} \frac{J_4 - J_3}{J_5}}.$$

For the alloy Be + Mg, for example, $T_m = 72^\circ \text{K}$ ($\epsilon = -1.77$, $\Delta(\text{Mg}) = 4.95 \times 10^{-3}$ eV, $\Theta(\text{Be}) = 950^\circ \text{K}$).

2. Let us consider the region of high temperatures. When $T \geq \Theta/2$ the integral with respect to ω in (13), which contains the temperature factor, becomes equal to

$$\frac{\hbar}{4kT} \int_0^\infty d\omega^2 \varphi(\omega^2) = \frac{\hbar}{4kT} \frac{1}{(1-\epsilon)}$$

Such an integral was calculated by Iosilevskii and Kagan^[21]. In other words, in this temperature region the scattering by the impurities makes a contribution to the line width, equal to

$$\frac{1}{T_1} = cr A_1^2 \frac{q_0^2}{2} \left(1 - \frac{\hbar q_0^2}{M} \frac{\hbar}{8kT} \frac{1}{(1-\epsilon)} \right). \quad (22')$$

It can be shown^[11] that at high temperatures the

integrals with respect to ω in the expressions for $T_1^{-1}(3)$ (formula (14)) and $T_1^{-1}(2)$ (formula (15)) become respectively equal to

$$J_1 = \frac{kT}{\hbar} \int_0^\infty \frac{d\omega^2}{\omega} \frac{1}{\pi} \frac{\gamma(\omega^2)}{\omega^2 - \omega_q^2} = \frac{1 - \Delta(\omega_q^2)}{2\omega_q^2} \frac{kT}{\hbar},$$

$$J_2 = \frac{2kT}{\hbar} \int_0^\infty \frac{d\omega^2}{\omega} \varphi(c, \omega) = \frac{kT}{\hbar} \frac{1}{\omega_q^2}.$$

Calculations of integrals of this type were performed by Kagan and Zhernov^[11]. Substituting these results in (14) and (15) we find that at high temperatures, for all values of ϵ , we get

$$\frac{1}{T_1(3)} = \frac{1}{T_{10}} = r \frac{kT}{M} \int |V_0(q)|^2 \frac{q^3}{\omega_q^2} dq, \quad (23)$$

$$\frac{1}{T_1(2)} = cr \frac{kT}{\hbar} \int \frac{a(q)q^3}{\omega_q^2} dq. \quad (24)$$

Let us consider now the case of a light impurity ($0 < \epsilon < 1$, $A_1 < A_0$). For small concentrations of impurities of this kind (see ≤ 10 at.%) at low temperatures we have

$$\frac{1}{T_1} - \frac{1}{T_{10}} \approx \frac{1}{T_1(3)}.$$

When $\epsilon > 0$ this quantity, as follows from (21), increases monotonically with temperature. We note that were it possible to select a solution such that $\epsilon A_0 < A_1$, then we would have $T_1^{-1}(2) > T_1^{-1}(3)$. In this case, in the low temperature interval, $T_1^{-1}(2)$ increases with increasing T and reaches a maximum value at $T = J_3 \Theta (2\epsilon J_4)^{-1}$. In the region $T \leq \Theta/2$, the temperature variation of that part of ΔH which depends on the impurity concentration is determined by relations (23) and (24).

In conclusion we note that if $V(q) = Aq$, then all the relations between the values of $T_1^{-1}(i)$, where $i = 1, 2, 3$, remain valid, and only the temperature behavior changes. It is easy to see that this reduces, apart from constant factors, to the following substitution in the presented expressions:

$$(T/\Theta)^n J_n \rightarrow \left(\frac{T}{\Theta}\right)^{n+2} J_{n+2}, \quad n > 0.$$

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