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1. Introduction

At low temperatires spins of a solid paramagnet relax to the equilibrium state toward the direct, one-phonon processes. (Van Vleck, 1941). This means the spin-flip is accompanied by creation or annihilation of a single phonon which frequency lies in the narrow band of frequencies. The width of this band is approximately equal to the width of resonance line $\Delta\omega$. Let us suppose that frequency band is centred around frequency which will be called further resonant frequency. All phonons with the frequencies from band $\Delta\omega$ will be called further resonant phonons. We suppose that all other lattice modes are in equilibrium.

If the spin-phonon interactions are more effective in comparison with the phonon-bath interaction we would expect a rise in the resonant phonons ''temperature'' (Van Vleck, 1941). In this case the relaxation time of the resonant phonons defines the time of returning of the full system to the equilibrium state. Such a situation is, following Van Vleck, termed a ''phonon-bath bottle-neck''. The main conclusion of Van Vleck was that the phonon-phonon interactions at low temperatures are not effective and, thus the interaction of the resonant phonons with the walls of the container or with the impurities gives a significant rise to relaxation of the full system.

There are many attempts to consider the theory of low-temperature paramagnetic relaxation, but the main Van Vleck conclusions were not changed. Various experimental techniques have been employed to demonstrate the existence of the bottle-neck. Some of them give the relaxation times (Scott et al., 1962; Nash, 1965; Standley et al., 1964), and others offer the possibility of a direct observation of the hot phonons (cf. Brya et al., 1972; Altshuler et al., 1972). However, in our opinion under some conditions the phonon-phonon interactions can play an essential role. We have in mind the fact that all previous calculations of the resonant phonons life-time were made in the collisionless regime. But it has been realized recently, that this life-time in the collision-dominated regime differs significantly from that in the collisionless regime. Since the life-time of thermal phonons depends on temperature it is possible that for a given frequency of the resonance ω_0 at liquid helium temperatures the collision-dominated regime occurs. In fact we prove for spin 1/2 that in the temperature interval, where usually the experiments have been made, (1 - 4 K) life-time due to the interaction of the resonant phonons with the walls of the container and life-time due to phonon-phonon interaction are of the same order.

Description of the nonequilibrium state of the spin system depends on what interaction is stronger: spin-spin or spin-lattice interaction. If the spin-spin interaction is greater than spin-lattice interaction the evolution of the system is described by Provotorov type equations. Otherwise, we must use the Bloch-type equations.

In order to develop the kinetic equations for our problem we use the Method of Nonequilibrium Statistical Operator (NSO) proposed by Zubarev (1971). Buishvili et al. (1972) have proved that this method is very convenient in application to the problem of the phonon bottleneck.

2. The Kinetic Equations

Let us consider a solid paramagnet in the resonance conditions. The general Hamiltonian of such a system has the form

$$\mathcal{H}(t) = \mathcal{H}_S + \mathcal{H}_{SS} + \mathcal{H}'_t + \mathcal{H}_{SL} + \mathcal{H}_L \equiv \mathcal{H} + \mathcal{H}'_t \ . \qquad (1)$$

The operator $\mathcal{H}_S$ describes the spectrum of the resonant levels. We consider such a spin system which has a system of energy levels with energy difference $\hbar\omega_0$. This system is well separated from other levels and lies at the bottom of the energy spectrum. The second term of (1) $\mathcal{H}_{SS}$ is the energy of the dipole-dipole interaction, $\mathcal{H}'_t$ represents the energy of the interaction of the spins with an ac magnetic field, $\mathcal{H}_{SL}$ is the energy interaction of spins with phonons and $\mathcal{H}_L$ is the lattice Hamiltonian. At low temperatures (liquid helium temperatures) the spin system is strongly coupled to the phonons of resonant frequency $\omega_Q \approx \omega_0$. The ac external magnetic field disturbs the spin system from the equilibrium state and subsequently leads to nonequilibrium distribution function of the resonant phonons. We suppose that all other phonons are in equilibrium. This is the reason to distinguish the resonant phonons from all other phonons.

$$\mathcal{H}_L = \mathcal{H}_Q + \mathcal{H}_{QT} + \mathcal{H}_T ,$$

where

$$\mathcal{H} = \sum_K \hbar\omega_K n_K \Delta(q,k) , \qquad (2)$$

is the Hamiltonian of the resonant phonons and

$$\Delta(q,k) = \begin{cases} 1 & if \quad |\omega_0 - \omega_{K+Q}| < \Delta\omega \\ 0 & if \quad |\omega_0 - \omega_{K+Q}| > \Delta\omega . \end{cases} \qquad (3)$$

The Hamiltonian of the interaction of the resonant and thermal phonons has the form

$$\mathcal{H}_{QT} = \sum_K (b_K V_K + V_K^+ b_K^+) \Delta(q,K) , \qquad (4)$$

where b_K^+, b_K are creation and annihilation operators of phonons, and V_K is equal to

$$V_K = 3 \sum_{K_1 K_2} V(K,K_1,K_2) A_{K_1} A_{K_2} (1-\Delta(q,K_1))(1-\Delta(q,K_2)),$$

$(K_1\ K_2, K_3)$ being the matrix element of three-phonon interaction and

$$A_K = b_K + b^+_{-K},$$

K denotes the pair of indices $K=(k,j)$, k is quasi-momentum vector, j -polarization and mode index.

Similar division of the phonon Hamiltonian is used in the sound attenuation problem (Maris, 1965; Paszkiewicz, 1972).

In what follows we shall consider a system with the resonant spectrum defined by Zeeman energy levels of the spins in external magnetic field H_0, thus $\mathcal{H}_S$ is equal to

$$\mathcal{H}_S = \mathcal{H}_Z = \hbar\omega_0 \sum_i S_i^z, \tag{5}$$

ω_0 is connected with H_0 - $\omega_0 = \gamma H_0$, γ being the gyromagnetic ratio. The spin interaction with ac magnetic field of the amplitude H_1 is taken in the form

$$\mathcal{H}'_t = \frac{\hbar\omega_1}{2} \sum_i (S_i^+ e^{-i\omega t} + S_i^- e^{i\omega t}), \quad \omega_1 = \gamma H_1. \tag{6}$$

Description of our system of the impurities in solids depends substantially on which of dipole-dipole interactions or spin-lattice interactions is greater. Firstly, we consider not too diluted systems in which the spins are more strongly coupled with themselves than with phonons. In such a case usually in the spin interactions the main role part of which is

$$\mathcal{H}_d^\circ = \frac{\hbar}{2} \sum_{i \neq j} A_{ij} (S_i^z S_j^z - \frac{1}{2} S_i^+ S_j^-). \tag{7}$$

We take the spin-lattice interaction $\mathcal{H}_{SL}$ in the following form

$$\mathcal{H}_{SL} = i\hbar \sum_{K,\alpha,j} g_K^\alpha S_j^\alpha (b_K e^{ikr_j} - b_K^+ e^{-ikr_j}), \quad (\alpha = +,-)$$

where after Sears et al. (Sears et al., 1967) we take

$$|g_K|^2 = g_K^+ g_K^- = N^{-1/2} \epsilon \left(\frac{\hbar\omega_0\omega_K}{12}\right)^{1/2},$$

where

$$\epsilon \; \alpha \; \begin{cases} \omega_0^{1/2} & \text{in the Kramers ion} \\ \omega_0^{-1/2} & \text{in the non-Kramers ion.} \end{cases}$$

Let us divide the spin-lattice interaction into two parts

$$\mathcal{H}_{SL} = \mathcal{H}'_{SL} + \mathcal{H}_{SQ} ,$$

where $\mathcal{H}'_{SL}$ is the great part

$$\mathcal{H}'_{SL} = i\hbar \sum_{K,\alpha,j} g_K^{\alpha} S_j^{\alpha} (b_K e^{iKr_j} - b_K^{+} e^{-iKr_j})(1 - \Delta(q,K)),$$

and the small part is the Hamiltonian of interaction of the resonance phonons with the spins

$$\mathcal{H}_{SQ} = i\hbar \sum_{K,\alpha,j} g_K^{\alpha} S_j^{\alpha} (b_K e^{iKr_j} - b_K^{+} e^{-iKr_j}) \Delta(q,K) . \quad (8)$$

It can be seen from Eq. (8) that the width of the resonance line $\Delta\omega$ defines this partition of $\mathcal{H}_{SL}$. Next we shall transform away the large part of $\mathcal{H}_{SL}$ (Frohlich, 1953). After canonical transformation in the second order in $\mathcal{H}_{SL}$, we obtain an effective spin-spin interaction the main secular part of which is equal to

$$\mathcal{H}^{\circ}_{eff} = \hbar \sum_{K,i,j} |g_K|^2 (1 - \Delta(q,K)) \{ S_i^{+} S_j^{-} \frac{e^{iK(r_i - r_j)}}{\omega_0 - \omega_K} - S_i^{-} S_j^{+} \frac{e^{iK(r_i - r_j)}}{\omega_0 + \omega_K} \} .$$

In contrast to dipole-dipole interaction the leading terms of $\mathcal{H}^{\circ}_{eff}$ for a long distance between ions have an oscillating character $\sin x / x^2$. It is possible that the phonon induced spin interaction can be larger than dipole-dipole interaction. Probably, such a case takes place in some organic complexes with Cu (Standley and Wright, 1964). In addition to effective spin interaction an effective spin-thermal phonon interaction of the type of (8) does appear. This leads to relaxation of the dipole subsystem. We can divide the Hamiltonian of the full system

$$\mathcal{H}(t) = \mathcal{H}_Z + \mathcal{H}_d + \mathcal{H}_{ind} + \mathcal{H}'_t + \mathcal{H}_Q + \mathcal{H}_T + \mathcal{H}_{SQ} + \mathcal{H}_{QT} + \mathcal{H}_T^{eff}$$

($\mathcal{H}_{ind}$ being the phonon induced spin-spin interaction, the secular part of which is $\mathcal{H}^{\circ}_{eff}$, and $\mathcal{H}^{eff}_{ST}$ is an effective spin-phonon interaction) into strong and weak parts. The strong part

$$\mathcal{H}_0 = \mathcal{H}_Z + \mathcal{H}^{\circ}_d + \mathcal{H}^{\circ}_{eff} + \mathcal{H}_Q + \mathcal{H}_T$$

describes the rapid returning to the internal equilibrium state of different subsystems - the coupled spin-resonant phonon subsystems and the thermal phonon subsystem.

The weak part

$$\mathcal{H}_{SQ} + \mathcal{H}_{QT} + \mathcal{H}^{eff}_{ST}$$

leads to the slow, smooth relaxation. With the interaction $\mathcal{H}^{\circ}_d + \mathcal{H}^{\circ}_{eff}$ there is connected a small time interval τ_d, such that after time much greater than τ_d the nonequilibrium state of the spin system is described by a set of average values of the operators P_m, which obey the following condition.

$$[P_m , \mathcal{H}_0] = \sum_n a_{mn} P_n . \qquad (9)$$

This condition guarantees that the nonequilibrium average values $\langle P_m \rangle^t$ apart from the free oscillations change slowly and are relevant to description of the nonequilibrium state of our system. In our first case there are four operators with such a property, namely $\mathcal{H}_Z, \mathcal{H}^{\circ}_d + \mathcal{H}^{\circ}_{eff}$, $\mathcal{H}_Q, \mathcal{H}_T$.

The operators P_m and thermodynamic parameters $F_m(t)$ coupled to $\langle P_m \rangle^t$ in the nonequilibrium thermodynamic sense define the entropy operator $S(t,0)$. In our case:

$$S(t,0) = \beta_Z(t)\, \mathcal{H}_Z + \beta_d(t)\, (\mathcal{H}^{\circ}_d + \mathcal{H}^{\circ}_{eff}) + \beta_Q(t)\, \mathcal{H}_Q + \beta \mathcal{H}_T + \phi(t) ,$$

where $\beta_Z(t)$, $\beta_d(t)$, $\beta_Q(t)$ are the parameters coupled to the $\langle \mathcal{H}_Z \rangle^t$, $\langle \mathcal{H}^{\circ}_d + \mathcal{H}^{\circ}_{eff} \rangle^t$, $\langle \mathcal{H}_Q \rangle^t$, respectively. The thermal bath has the temperature T and β is equal to familiar $(kT)^{-1}$. With the help of this entropy operator we can introduce the local equilibrium statistical operator $\rho_\ell(t,0)$

$$\rho_\ell(t,0) = \exp\{-S(t,0)\}, \qquad Tr\rho_\ell(t,0) = 1,$$

$\phi(t)$ is the normalization factor of $\rho_\ell(t,0)$. Using the NSO method we construct the nonequilibrium statistic operator, $\rho(t,0)$ being the functional of $\rho_\ell(t,0)$ relevant to the given stage of evolution

$$\rho(t,0) = \epsilon \int_{-\infty}^{0} dt_1 \, e^{\epsilon t_1} \; U^{+}(t+t_1, t_1) \, \rho_\ell(t+t_1, 0) \, U(t+t_1, t_1) \,, \tag{10}$$

where

$$U(t,t') = \exp\left(-\frac{i}{h}\mathcal{H}_0 t\right) T \exp\left\{\frac{1}{ih}\int_{t'}^{t} \mathcal{H}'_{t_1}(t_1)\, dt_1\right\},$$

$$\mathcal{H}'_{t_1}(t\ ) = \exp\left(\frac{i}{h}\mathcal{H}_0 t_1\right) \mathcal{H}'_{t_1} \exp\left\{-\frac{i}{h}\mathcal{H}_0 t_1\right\}.$$

After taking the thermodynamic limit, the next step is to take ϵ_{-}. Averaging the operator equations of motion for P'_m s over the nonequilibrium statistic operator (10) we obtain the energy balance equations, i.e. kinetic equations for $\langle\mathcal{H}_Z\rangle^t$, $\langle\mathcal{H}^{\circ}_d + \mathcal{H}^{\circ}_{eff}\rangle^t$, and $\langle\mathcal{H}_Q\rangle$. We compute the coefficients of these equations with an accuracy to terms of the second order in all interaction, i.e. $\mathcal{H}'_t$, $\mathcal{H}_{QT}$, $\mathcal{H}_{ST}$ and $\mathcal{H}^{eff}_{ST}$. The balance equations can be rewritten in the form of equations for ''reverse'' Zeeman $\beta_Z(t)$, dipole $\beta_d(t)$ and resonant phonon temperatures $\beta_d(t)$ (cf. Buchbinder and Kessel, 1972). In the high-temperature limit we get

$$\frac{d\beta_Z(t)}{dt} = -W\left(\beta_2(t) + \frac{\omega-\omega_0}{\omega_0}\beta_d(t)\right) - \frac{\beta}{\beta_Q(t)}\,\frac{\beta_Z(t)-\beta_Q(t)}{T_{ZQ}}\,, \tag{11a}$$

$$\frac{d\beta_d(t)}{dt} = -W\frac{(\omega-\omega_0)\,\omega_0}{\omega_d^2}\left(\beta_Z(t) + \frac{\omega-\omega_0}{\omega_0}\beta_d(t)\right) - \frac{\beta}{\beta_Q(t)}\,\frac{\beta_d(t)-\beta_d(t)}{T_{dQ}}\,, \tag{11b}$$

$$\frac{d\beta_Q(t)}{dt} = -\frac{\beta_Q(t)}{\beta}\,\frac{\beta_Q(t)\cdot\beta_Z(t)}{T_{QZ}} - \frac{\beta_Q(t)}{\beta}\,\frac{\beta_Q(t)-\beta_Q(t)}{T_{Qd}} - \frac{\beta_d(t)}{\beta}\,\frac{\beta_Q(t)-\beta}{T_{QT}}\,. \tag{11c}$$

Now, we shall explain our notations. The main quantity is the function of the dipole-dipole line shape $f(\omega)$ defined as the Fourier transform of the spin correlation function

$$f(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{i\omega t}\, f(t) \,, \qquad f(t) = \frac{Tr_S\, S^+ S^-(t)}{Tr_S\, S^+ S^-} .$$

Some other quantities are functions of $f(\omega)$

$$W = \pi\, \omega_1^2\, f(\omega - \omega_0) \,, \qquad h^2\, \omega_d^2 = \frac{Tr_S\, \mathcal{H}_d^2}{Tr_S\, S_z^2} .$$

The kinetic coefficients T_{ZQ}, T_{QZ}, T_{QT}, T_{Qd}, T_{dQ} are equal to

$$T_{ZQ}^{-1} = \frac{12\pi}{4 h\, \omega_0 \beta} |g_Q|^2\, \rho(\omega_0) \,, \qquad T_{dQ}^{-1} = T_{ZQ} \frac{\Delta_2 + \omega_0^2}{\omega_d^2} \,,$$

$$T_{QZ}^{-1} = \frac{4\pi N S(S+1)\beta}{3\Delta\omega} |g_Q|^2\, h\omega_0 \,, \qquad T_{Qd}^{-1} = T_{QZ}^{-1} \frac{\Delta_2 + \omega_0^2}{\omega_d^2} \,,$$

$$T_{QT}^{-1} = \frac{J_{V_Q^+ V_Q}(-\omega_0)}{h^2 (1 + \bar{n}(\omega_0))} \,,$$

where N is the number of paramagnetic impurities in the sample and Δ_2 is the second moment of the line-shape-function. The average value of the number of the resonant phonons in equilibrium state with temperature of thermal bath T is equal to $\bar{n}(\omega_Q) = (\exp(\beta h \omega_Q) - 1)^{-1}$.

The relaxation time of resonance phonons is connected with $J_{V_Q^+ V_Q}(\omega)$ the spectral density of phonon correlation function defined as

$$\langle V_Q^+ V_Q(t) \rangle = \int_{-\infty}^{\infty} d\omega\, e^{-i\omega t}\, J_{V_Q^+ V_Q}(\omega) .$$

The bracket <...> denotes equilibrium average

$$<\ldots> = Tr\,(\ldots \frac{e^{-\beta H_T}}{Tr \exp\{-\beta H_T\}}) \, .$$

In agreement with the result of Van Vleck (1941) and Orbach (1961) we obtain the spin-lattice relaxation time proportional to temperature

$$T_{ZQ}^{-1} \approx T \, .$$

In the equations (11) we omit relaxation times due to the effective spin-thermal bath interaction. In the opposite case where the dipole interaction is much weaker than spin-phonon interaction the main part of Hamiltonian $\mathcal{H}_0$ is equal to

$$\mathcal{H}_0 = \mathcal{H}_Z + \mathcal{H}_Q + \mathcal{H}_T \, .$$

Now, the set of operators which satisfies the condition (9) consists of $\mathcal{H}_T$, $\mathcal{H}_Q$, $S_\alpha = \sum_{i=1} S_i^\alpha (\alpha = x, y, z)$. $\mathcal{H}_{SQ}$,

$\mathcal{H}_{QT}$ cause slow smooth relaxation. For the average values of S^α we obtain modified Bloch equations. For simplicity we consider the spin one-half case only. This allows us to compare the asymptotic time behaviour for systems governed by equations of Provotorov and Bloch-type. Using the same method as that for derivation of Provotorov-type equations we obtain modified Bloch equations.

$$\frac{d<S_Z>^t}{dt} = - A\{(1 + 2<n_Q>^t\,)\,<S_Z>^t - \frac{N}{2}\} + \omega_1 \cos\omega t <S_y>^t \, , \tag{12a}$$

$$\frac{d<S_x>^t}{dt} = - \frac{A}{2}\{1 + 2<n_Q>^t\}<S_x>^t - \omega_0 <S_y>^t \, , \tag{12b}$$

$$\frac{d<S_y>^t}{dt} = - \frac{A}{2}\{1 + 2<n_Q>^t\}<S_y>^t + \omega_0 <S_x>^t - \omega_1 \cos\omega t <S_Z>^t \, , \tag{12c}$$

$$\frac{d\langle n_Q\rangle^t}{dt} = \frac{A}{\rho(\omega_0)\,\Delta\omega}\{(1+2\langle n_Q\rangle^t)\langle S_z\rangle^t - \frac{N}{2}\} - \frac{1}{T_{QT}}(\langle n_Q\rangle^t - \bar{n}(\omega_0)) \tag{12d}$$

where

$$A = 2\pi\,|g_Q|^2\,\rho(\omega_0)\ .$$

In both systems of equations (11) and (12) we discard all terms describing the relaxation of the spin subsystem direct to thermal bath. These terms are smaller than the terms due to resonance phonons.

3. Phonon Relaxation Time

The relation between the frequency ω_0 and life-time of thermal phonons τ_{th} influences very simply the result of computations of the relaxation time T_{QT}. Due to the nature of Bose-Einstein statistics such thermal phonons with the energy of the order of kT play an essential role in the dynamics of the phonon system. In the collisionless regime $(\omega_0\,\tau_{th} \gg 1)$ there are a few collisions during the characteristic time ω_0^{-1} and interactions between the thermal phonons result in a sort of the self-consistent field. Therefore, thermal phonons never reach their equilibrium. In this regime the relaxation-time of resonant phonons can be found from the probability of transition of the acoustic phonons (or from suitable limit of phonon mass operator). Following Kwok (1967) we decouple the correlation function $\langle V_Q^+ V_Q(t_1)\rangle$ in the product of two correlation functions of the type of $\langle A_K A_K^+(t_1)\rangle$. In such a manner we obtain

$$T_{QT}^{-1} = 36\pi \sum_{K_1,K_2\neq Q} |V(K_1,K_2,Q)|^2 \int_{-\infty}^{\infty} d\Omega_1\, d\Omega_2\, J_{K_1}(\Omega_1)\, J_{K_2}(\Omega_2)\ \times$$

$$\times\,(1+\bar{n}(\Omega_1)+\bar{n}(\Omega_2))\ \delta(\Omega_1+\Omega_2-\omega_Q)\ .$$

The spectral density function $J_K(\Omega)$ is defined with the help of the phonon Green function (Kwok, 1967).

$$J_K(\Omega) = i(\langle\langle A_K ; A_K^{+} \rangle\rangle_{\Omega+i\epsilon} - \langle\langle A_K ; A_K^{+} \rangle\rangle_{\Omega-i\epsilon}) .$$

If we use the harmonic approximation for the spectral density we obtain the standard Van Vleck (1941) result, (usually called Landau-Rumer formula).

$$T_{QT}^{-1} \; a \, \omega_0 \, T^4 .$$

In the opposite case, i.e. in the collision-dominated regime there are many collisions during the characteristic time ω_0^{-1}, the thermal phonons collide frequently and relax toward a local equilibrium state. The suitable equations describing such a local equilibrium are some kind of the Bottzmann equations. Such equations were developed by many authors but their suitable derivation for our purpose is that of Sham (1967).

In order to obtain the spectral density function $J_{V_Q^+ V_Q}$ we must solve the Boltzmann like equations by Sham (1967). We do this following Niklasson (1972) in the relaxation-time approximation. In such an approximation in the theory there appear two parameters: the relaxation time connected with the normal processes τ_N, and that connected with the momentum nonconservating (Umklapp) processes τ_U. Solving the Boltzmann-like equations for cubic crystals in the diffusion region, where the normal and Umklapp processes are equally frequent $(\omega_0 \tau \ll 1 \; , \omega_0 \tau_U \ll 1 \; , \tau^{-1} = \tau_N^{-1} + \tau_U^{-1})$ we get

$$T_{QT}^{-1} = \left(\frac{c_p}{c_v} - 1\right) \frac{\omega_0^2 (c_{||} \, q)^2 \tau_U}{\omega_0^2 + [(c_{||} \, q)^2 \, \tau_U]^2} , \qquad (13)$$

$Q = (q,$ longitudinal acoustic). c_p being the heat capacity per unit volume at constant pressure, c_v at constant volume, $c_{||}$ is the second sound velocity approximately equal to $c/\sqrt{3}$, where c is the sound velocity which we take the same for differently polarized acoustic oscillations. In the presence of the impurities of low concentrations we should replace relaxation time τ_u by that τ_R connected with all resistive processes.

For great concentrations of impurities we can use approximate results of Miller (1965).

The Boltzmann-like equations admit a solution describing the propagation of the wave-second sound mode. But in the considered systems such a mode is strongly damped due to the interactions with the impurities which change the momentum of phonons. Probably only in samples used by Standley et al. (1964), which are stoichiometric crystals, this mode can be propagated.

4. Discussions

In this section we consider the process of relaxation in the system of the fictious spins 1/2 after turning off a disturbing field. The equation for temperature (11) are very complicated nonlinear equations. It is a hard task to find the solution of these equations. Recently, Bushvili et al. (1971) have solved equations of this type for some simple cases. If we drop all dipolar terms in balance equations for $\langle \mathcal{H}_Z \rangle^t$, $\langle \mathcal{H}_Q \rangle^t$ we obtain the nonlinear equations of Brya and Wagner (1967). However, this approximation is obviously incorrect. The discussion of our equations for temperatures in the presence of dipolar terms is very complicated thus we shall consider the role of phonons in the low-temperature paramagnetic relaxation without taking into account the dipolar terms. For this purpose we shall use our modified Bloch equations. For the long time in the lowest order in deviation from equilibrium the equations for $\langle S_x \rangle^t$, $\langle S_y \rangle^t$ are decoupled from equations for $\langle S_Z \rangle^t$ and $\langle n_Q \rangle^t$.

In agreement with the Mims and Taylor (1971) the equations for $\langle S_Z \rangle^t$ and $\langle n_Q \rangle^t$ coincide with those of Brya and Wagner (1967). These equations can be written in terms of the following quantities used by Brya and Wagner

$$u(t) = \frac{n_- - n_+}{n_+^\circ - n_-^\circ}, \quad y = \frac{\langle n_Q \rangle^+ - \bar{n}(\omega_Q)}{\bar{n}(\omega_Q) + 1/2},$$

where n_+, n_- are the numbers of ions in the lower and the upper state, respectively; n_+° , n_-° are the same numbers but in equilibrium state,

$$\frac{du}{dt} = -\frac{1}{T_{ZQ}}(u + 1 + uy) ,$$

$$\frac{dy}{dt} = \frac{1}{T_{QT}}(\sigma(u + 1) + \sigma uy - y) ,$$

T_{ZQ} being the direct spin-lattice rate at the ambient temperature

$$T_{ZQ}^{-1} = A(1 + 2\bar{n}(\omega_0)) ,$$

and σ is the "bottle-neck factor" equal to the energy exchange rate between spins and phonons divided by energy exchange rate between phonons and bath

$$\sigma = \frac{T_{QT}}{T_{ZQ}} \frac{h\omega_0 (n_+^\circ - n_-^\circ)}{h\omega_0 \Delta\omega \rho(\omega_0)(1 + \bar{n}(\omega_0))} .$$

Brya and Wagner (1967) have discussed eqs. (12a, 12d) in details. They have shown that these equations describe the phonon avalanche <u>if</u> the spin temperature is initially negative and resonant phonons cannot be immediately absorbed by the thermal bath.

Although the rate equations are nonlinear and can be solved only numerically, it is possible to find the solutions for certain special cases. From the point of view of NSO method we are interested in the behaviour of the system near the end of decay, where $u \to -1$, $y \to 0$, $\langle S_x \rangle^t \to 0$, $\langle S_y \rangle^t \to 0$. Following Brya and Wagner (1967), we obtain two decay rates, the slower of which is

$$T_{1A} = (6 + 1)\ T_{ZQ} \, . \qquad (14)$$

This quantity is the asymptotic time constant obtained in pulse saturation studies of Scott and Jefferies (1962); Ruby et al. (1962); Nash (1965), Standley et al. (1965).

Finally, we shall consider the role of phonon-phonon interactions in the bottle-neck. Following Guyer and Krumhansl (1966) we divide the temperature interval into two parts; below and above the temperature of conductivity peak $T_M \approx 0.025\ T_D$, where T_D is the Debaye temperature of the crystal. For mostly used frequencies of the order of 10^{10} $\sec^{-1}$ according to Guyer and Krumhansl (1966) the system is found in the diffusion *region and below T_M in the collisionless one.* In the collisionless regime $T_{QT} \propto T^{-4}$ and in high-temperature approximation we get

$$T_{1A} \propto \left(\frac{1}{BT^6} + \frac{1}{cT} \right) .$$

From this formula one draws usually a conclusion that for low temperatures the phonon-phonon interactions are much less effective than with the walls or impurities (Stoneham, 1965). However, this statement is valid only *in the high-temperature approximation, i.e.* $\hbar\omega_0 \ll kT$.

As we have shown in sect. 3 temperature and field dependence of T_{QT} differ significantly in the collisionless and collision-dominated regimes. It makes it possible that for substances with sufficiently low Debye temperatures T_D , the system at liquid helium temperatures can be found in the diffusion region. The Debye temperature in this case should be of the order of 60° K.

Let us estimate the ratio of σ_W due to the interactions of phonons with the walls and σ_d for phonon-phonon interactions in the diffusion region. We assume the following crude figures for evaluating the order of magnitude of formulae: following Niklasson (1972) we take $\tau_U \approx 10^{-9}$ sec, $\omega \approx 10^{9}$ $\sec^{-1}$ and time of flight of phonons across the sample equal to 10^{-6} sec (the size of sample 0.25 cm), then

$$\frac{\sigma_W}{\sigma_d} \; a \left(\frac{c_p}{c_v} - 1 \right) 10^4 .$$

This result is valid only for temperatures above T_u. Both approximations - cubic crystals symmetry and one branch dispersionless model cause the presence of the small parameter $(c_p/c_v - 1)$. This parameter is of the order of 10^{-4} at $T \approx 10$°K (Guyer and Krumhansl, 1966); Bleaumont et al. (1961)). Thus the ratio of σ_W and σ_d is of the order of unity.

Recently Brya et al. (1972) have really observed the hot phonons in Ni -doped MgO using the Brillouine light scattering. The Debye temperature for MgO is 700°K and the diffusion region appears above $T_M = 19$°K. From the result of Brya et al. (1972) it follows that the bottleneck persits up to 40°K, but the hot phonon temperature is constant up to temperature of 16°K. Above this temperature up to 40°K the number of hot phonons decreases to zero. In our opinion up to 16°K the phonon system is found in the collisionless region and according to usual formulas the temperature of resonant phonons changes very slowly. Above the temperature of 16°K the collision-dominated region occurs and the resonant phonon temperature quickly relaxes toward equilibrium. Hence, it seems reasonable to expect that for direct processes the phonon-phonon interactions can contribute the frequency and temperature-dependent mechanism of disappearing of bottle-neck, although other phonon processes, e.g., Raman processes, can be prevailing at elevated temperatures. The measurements in wide region of frequencies and temperatures may provide the proof of this suggestions.

5. Conclusions

The aim of the study was to consider the role of phonon-phonon interactions in the low-temperature paramagnetic relaxation. If dipole-dipole interactions are greater than spin-lattice interactions we obtain in rigorous manner the system of equations for reverse temperatures of Zeeman, dipolar and resonant phonon subsystem. In the opposite case we have developed the modified Bloch equations. We have shown that the asymptotic solutions of this equations coincide with those obtained by Brya and Wagner (1967). Using this solution we have revised the influence of phonon interactions on the low-temperature paramagnetic relaxation. If we have found our system in the hydrodynamic region thermal phonons can form the thermal bath and for great dimensions of samples the influence of surface can be neglected. Probably these conditions took place in experiments of Brya et al. (1971).

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