

Resonant-radiation transfer and nonequilibrium phonons in ruby

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The usual Biberman–Holstein theory of resonant-radiation transfer is valid only when the reradiating center “forgets,” after absorbing the radiation quantum, everything but the fact that it is excited. This is possible if the center interacts strongly with the “thermostat.” In a number of cases, for example for resonant phonons in ruby, this is not the case. It is also possible to formulate an alternative transfer theory, and this theory is developed in this paper.

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INTRODUCTION

An alternative to the known quantum theory of Biberman and Holstein^[1, 2] is developed here for resonant-radiation transfer. The present work was stimulated by experiments on nonequilibrium phonons in ruby.^[3, 7] We shall therefore deal henceforth with phonons and two-level impurity centers (rather than photons and atoms).

The Biberman–Holstein equation, the derivation of which will be analyzed in Sec. 1, presupposes that a strong interaction exists between the impurity centers and the “thermostat.” This is precisely why it suffices, for a description of the nonequilibrium state of the system of centers, to know only the concentrations N_1 and N_2 of the centers at the lower level 1 and at the upper level 2; all other nonequilibrium parameters “are forgotten” in the course of interaction with the thermostat.

On the other hand if the system of centers interacts only with resonant radiation, then the description of its nonequilibrium state is much more complicated—the system “remembers” better the excitation conditions, and more parameters are needed for its description. Thus, for example, the excitation energy of the system of centers is described in the Biberman–Holstein equations in integral fashion—by the parameter N_2 . If, however, the system remembers the excitation conditions, then its description calls for knowledge of the spectral density $F(\omega)$ of the center excitation energy (ω is the center excitation energy reckoned from the level $E_1 = 0$). The total number, N_2 , of the excited centers, is obtained as an integral of $F(\omega)$ with respect to the frequencies. Analogously, instead of the phonon distribution function in the momenta n_q there appears the spectral density of the phonon field $s(\omega, q)$, which is proportional to the degree of excitation of the mode q at the frequency ω . The fact that it becomes necessary to speak of the excitation of the q mode not at the natural frequency ω_q but at an arbitrary frequency ω means that the phonon field executes forced rather than natural oscillations.

The spectral densities $F(\omega)$ and $s(\omega, q)$ are closely related with the Green’s functions for the centers and phonons. The interaction of resonant radiation with two-level centers was investigated in terms of Green’s

functions in a number of papers.^[8–14] In all these studies, however, the formulation of the problem was the following: if at the initial instant of time there is one excited center or one radiation quantum, how will the system evolve subsequently? Unfortunately, the solution of this problem does not answer the questions that arise in the experiment.

We therefore obtain in the present paper a system of equations for the functions $F(\omega, r, t)$, which makes it possible to determine the factually measurable quantities for a given spatial distribution and time dependence of the excitation of a system of centers and phonons. Concrete problems will be considered separately.

1. THE BIBERMAN-HOLSTEIN THEORY

The original radiation transfer equation derived by Biberman and Holstein is formulated as an equation for $N_2(r, t)$. We present for this equation a somewhat different derivation that contains the phonon distribution function $n_q(r, t)$ of interest to us. This is done for two reasons: first, to make clear the assumption on which the derivation is based and, second, to be able to compare the equations for N_2 and n_q with the equations we obtain for $F(\omega)$ and $s(\omega, q)$.

The main premises on which the Biberman–Holstein equations are based are the following. For a complete description of the state of the system it suffices to know the phonon momentum distribution function n_q , the concentration N_1 of the centers located at the lower level 1, and the concentration N_2 of the centers on the upper level 2. All these quantities (n_q, N_1 , and N_2) can depend on the position in space r and on the time t .

The probability that a phonon with momentum q (and energy ω_q) will be absorbed is assumed to be

$$A^a N_2 \varphi^a(\omega_q), \quad (1.1)$$

where A^a is a certain constant, and the function $\varphi^a(\omega)$ determines the absorption line shape, it is convenient to choose it such that $\varphi^a(\omega) = 1$ at the center of the absorption line, i.e., at $\omega = \omega_0^a$. The absorption line width is then

$$\delta\omega^a = \int d\omega \varphi^a(\omega). \quad (1.2)$$

The phonon-production probability is assumed to be

$$A^a N_2 \varphi^a(\omega_q) (n_q + 1), \quad (1.3)$$

where A^e is a new constant, while the function $\varphi^e(\omega)$, normalized in analogy with $\varphi^a(\omega)$, describes the emission line shape.

Under these assumptions, the equation for the phonons takes the form

$$\left(\frac{\partial}{\partial t} + v_q \nabla \right) n_q = C_q \{N_1, N_2, n_q\} + S_q, \quad (1.4)$$

where

$$C_q \{N_1, N_2, n_q\} = -A^a \varphi^a(\omega_q) N_1 n_q + A^e \varphi^e(\omega_q) N_2 (n_q + 1), \quad (1.5)$$

and the second term in the right-hand side of (1.4) is responsible for the phonon-phonon interactions.

Since each phonon absorption act is accompanied by a transition $1 \rightarrow 2$, and each emission act by a transition $2 \rightarrow 1$, we can easily write down the kinetic equations for N_1 and N_2 :

$$\frac{\partial N_1}{\partial t} = \int \frac{d^3 q}{(2\pi)^3} C_q \{N_1, N_2, n_q\} - \frac{1}{\tau_1} N_1 + G_1, \quad (1.6)$$

$$\frac{\partial N_2}{\partial t} = - \int \frac{d^3 q}{(2\pi)^3} C_q \{N_1, N_2, n_q\} - \frac{1}{\tau_2} N_2 + G_2. \quad (1.7)$$

Here G_1 and G_2 describe the pumping of the levels E_1 and E_2 by external sources, while τ_1 and τ_2 describe their depletion which is not connected with resonant phonons (for example in the $E_1 \rightarrow E_0$ and $E_2 \rightarrow E_0$ transitions to the lower level E_0).

Let us discuss the premises on which the foregoing equations are based. The most serious is the assumption (1.3) concerning the distribution of the phonons emitted by the excited centers. It means that the spectrum of the phonons radiated by the system of centers depends neither on the system state nor on its prior history, while the total number of emitted phonons depends on a single parameter N_2 .

We emphasize first of all that in the Biberman-Holstein theory the width $\delta\omega^e$ cannot be due to the natural width τ^{-1} , since an isolated center, as is known from the theory of resonant fluorescence,^[15] forgets the prior history only in the case of broadband excitation. The width $\delta\omega^e$ can be due to the scatter of the energies of the $E_2 \rightarrow E_1$ transition for different centers (inhomogeneous broadening) or to splitting of the transition $E_2 \rightarrow E_1$ at one center (vibrational structure). In either case the assumption (1.3) means that the relative probabilities of filling different states of the "level" 2 are fixed, although the absolute ones are arbitrary and are determined by the parameter N_2 . This is possible only if the system of centers exchange intensively energies of the order $\delta\omega^e$ with the thermostat, and the distribution over the states of the level 2 is determined by the thermostat. The arbitrariness of N_2 means that exchange of high energies is difficult, therefore the interaction with the thermostat does not change the total number of excited centers.

One might think that the role of the interaction with the thermostat can be played also by an interaction V between centers, which satisfies the condition $\delta\omega^e \ll V \ll \omega_0 \equiv E_2 - E_1$. This interaction does indeed conserve the number of excited centers and establishes some equilibrium between them. This interaction, how-

ever, conserves not only the number of the excited centers but also their energy. Therefore even the relative occupation numbers of the states of the level 2 should remember the excitation conditions.

The situation is the same with the assumption (1.1).

If we assume that Eqs. (1.4), (1.6), and (1.7) are valid also for the described states which are closed to thermodynamic equilibrium, then the parameters of the emission and absorption lines should coincide:

$$A^a = A^e = A, \quad \varphi^a(\omega) = \varphi^e(\omega) = \varphi(\omega), \quad \delta\omega^a = \delta\omega^e = \delta\omega. \quad (1.8)$$

In fact, under thermodynamic equilibrium we have $C_q = 0$. It is easy to verify that this identity is satisfied at temperatures $T \gg \delta\omega^e$ only if (18) holds.

The constant A^e can be expressed in terms of the probability τ^{-1} of the spontaneous emission of the phonon. The number of phonons produced in 1 cm³ per second in spontaneous emission is

$$\int \frac{d^3 q}{(2\pi)^3} \varphi^e(\omega_q) A^e N_2 = A^e N_2 \rho_0 \delta\omega^e, \quad (1.9)$$

where ρ_0 is the phonon-state density corresponding to the line center

$$\rho_0 = \rho(\omega_0), \quad \rho(\omega) = \int \frac{d^3 q}{(2\pi)^3} \delta(\omega - \omega_q). \quad (1.10)$$

It is clear from (1.9) that

$$\tau^{-1} = A^e \rho_0 \delta\omega^e. \quad (1.11)$$

The gist of the last remark, which pertains to the region of applicability of the system (1.4), (1.6), and (1.7), is the following. Since this system makes use of the phonon distribution function n_q , this means that the width of the phonon distribution should exceed the reciprocal absorption time, i.e.,

$$\Delta\omega_q \gg \tau^{-1} = A^e N_1. \quad (1.12)$$

In many cases the depletion of the lower level is negligible, i.e., $N_2 \ll N_1$. Then, adding (1.6) and (1.7), we have

$$\frac{\partial}{\partial t} N_1 = - \frac{1}{\tau_1} N_1 + G_1 + G_2. \quad (1.13)$$

This equation yields $N_1 \equiv N^*(\mathbf{r}, t)$ and we are left with two equations for N_2 and n_q . If furthermore $n_q \ll 1$, then these equations can be written in the form

$$\left(\frac{\partial}{\partial t} + v_q \nabla \right) n_q = -N^* A^a \varphi^a(\omega_q) n_q + A^e \varphi^e(\omega_q) N_2 + \hat{S} n_q, \quad (1.14)$$

$$\frac{\partial}{\partial t} N_2 = - \frac{1}{\tau_2} N_2 + N^* A^e \int \frac{d^3 q}{(2\pi)^3} \varphi^e(\omega_q) n_q + G_2 - \frac{1}{\tau_2} N_2. \quad (1.15)$$

If we neglect the phonon-phonon interactions (this can always be done for photons), then we can use (1.14) to express n_q in terms of N_2 . Substituting this in (1.15), we obtain an equation for N_2 ; if we neglect retardation in this equation, i.e., the time of ballistic flight of the phonon through the active volume (this can always be done for photons), we obtain the well-known Biberman-Holstein equation.

2. GENERAL EQUATIONS

In this section we derive the equations for the propagation of excitation in a system of randomly disposed

two-level centers that interact with the phonon field. The reason why the system is not at equilibrium is that the centers are optically pumped also under the influence of the phonon momentum injected on the crystal boundary.

The nonstationarity and spatial inhomogeneity of the system are determined by the excitation conditions, i.e., by the geometric dimensions of the excited regions and by the duration of the excitation (or by the duration of the relaxation of the consequences of this excitation).

Owing to the microinhomogeneity of the crystal, the centers located at one macroscopic point \mathbf{r} can have several different energies of the $E_2 - E_1$ transition. Writing this in the form $E_2 - E_1 = \omega_0 + \nu$, where ω_0 is the average transition energy, we obtain the parameter ν that characterizes the center. The distribution of the centers in ν is given by the function $\phi(\nu)$, normalized by the condition

$$\int d\nu \phi(\nu) = 1, \quad \phi(0) = (\Delta\omega_0)^{-1}. \quad (2.1)$$

The quantity $\Delta\omega_0$ can be called the inhomogeneous broadening.

Bearing applications to ruby in mind, it must be recognized that the levels E_2 and E_1 are in fact Kramers doublets (see Figs. 1 and 2). The degeneracy of these levels can be lifted by an external magnetic field or by the random magnetic fields of neighboring centers. Therefore the magnetic field H at the location of the center serves, in addition to ν , as a parameter that characterizes the center. The components of each doublet will be marked by an additional index \pm (spin orientation), and their splittings will be designated $2\delta_1$ and $2\delta_2$.

The state of the system of centers is described by Green's functions of the following type:

$$G_{ss'}^{\pm\pm}(\mathbf{R}, \mathbf{R}'; t_1, t_2) = \langle a_{s\mathbf{R}}^\pm(t_1)^+, a_{s'\mathbf{R}'}^\pm(t_2)^- \rangle,$$

where $(a_{s\mathbf{R}}^\pm)^*$ is the operator of electron production on a

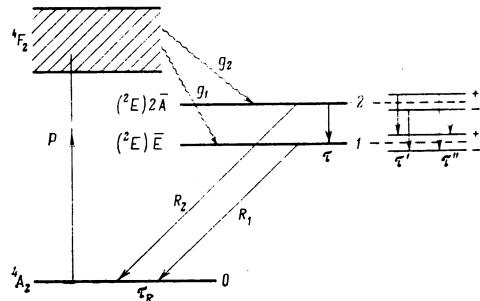


FIG. 1. Levels of Cr^{3+} ion in Al_2O_3 and transitions between them. P—optical pumping from the ground level ${}^4\text{A}_2$ to the absorption band ${}^4\text{F}_2$. The populations of the metastable levels $({}^2\text{E})\bar{E}$ and $({}^2\text{E})2\bar{A}$ in nonradiative transitions are g_1 and g_2 . Luminescence corresponds to the transitions R_1 and R_2 (radiative time τ_R). The figure shows also the transition between metastable levels with emission of a phonon (time τ). In the right-hand side of the figure is shown the structure of the Kramers doublets \bar{E} and $2\bar{A}$ in a magnetic field and the transitions between them without spin flip (time τ') and with spin flip (time τ'').

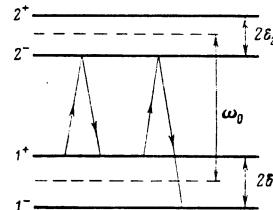


FIG. 2. Reradiation of spectral density. On the left is shown one of the four reradiation processes without spin flip: $1^+ \rightarrow 2^+ \rightarrow 1^+$; the remaining processes of this type are $1^+ \rightarrow 2^- \rightarrow 1^+$ and $1^- \rightarrow 2^- \rightarrow 1^-$. On the right is shown one of the four reradiation processes with spin flip: $1^+ \rightarrow 2^- \rightarrow 1^-$; the remaining processes of this type are $1^+ \rightarrow 2^+ \rightarrow 1^-$ and $1^- \rightarrow 2^+ \rightarrow 1^+$. The four processes with spin flip correspond to the four terms in the kernel (3.23).

center located at the point \mathbf{R} , in a state $s=1$ or 2 and with corresponding spin orientation. We shall assume that the states of the different centers are not correlated, i.e., $G=0$ if $\mathbf{R} \neq \mathbf{R}'$, and that the centers are excited "incoherently," i.e., only the "diagonal" functions $G_{ss}^{++} = G_s^+$ and $G_{ss}^{--} = G_s^-$ differ from zero. We assume next that the inhomogeneity and the nonstationarity of the system are weak. Therefore the dependence on the discrete \mathbf{R} in the Green's function $G_s^*(\mathbf{R}; t_1, t_2)$ is transformed into a dependence on the macroscopic coordinate \mathbf{r} . The macroscopic time will be $t = \frac{1}{2}(T_1 + T_2)$, and it is natural to take the Fourier transform with respect to the difference $t_1 - t_2$ and change over to the variable ε . Thus the functions $G_s^*(\mathbf{r}, t; \varepsilon)$ figure in the theory. In view of the foregoing, these functions depend also on the parameters ν and H that characterize the "type" of the center.

The equations for these functions are derived with the aid of the Keldysh diagram technique^[16]; it is convenient to transform these equations into generalized kinetic equations.^[17] The generalized kinetic equations are functions of the generalized occupation numbers $n_s^*(\nu, H; \varepsilon)$, the level widths $\Gamma_s^*(\nu, H; \varepsilon)$, and the level shifts $\Delta E_s^*(\nu, H; \varepsilon)$. The Green's functions are replaced by the generalized occupation numbers $n(\omega, \mathbf{q})$, by the widths $\gamma(\omega, \mathbf{q})$, and by the shifts $\Delta\omega(\omega, \mathbf{q})$. All the occupation numbers, widths, and shifts depend on \mathbf{r} and t . The broadenings and the shifts of the levels lead to the appearance of broadened and shifted delta functions; for examples, for photons we get in place of $\delta(\omega - \omega_0)$

$$\Delta(\omega, \mathbf{q}) = \frac{\gamma(\omega, \mathbf{q})}{2\pi} \left\{ [\omega - \omega_0 - \Delta\omega(\omega, \mathbf{q})]^2 + \left[\frac{1}{2}\gamma(\omega, \mathbf{q}) \right]^2 \right\}^{-1}. \quad (2.2)$$

The functions $\Delta_s^*(\varepsilon)$ for the centers are determined in similar fashion.

Having made these remarks, we can write down the generalized kinetic equations on the basis of their formal analogy with the ordinary kinetic equations.^[17,18]

The balance equations for the centers is

$$\Delta_s^*(\varepsilon) - \frac{\partial}{\partial t} \Delta_s^*(\varepsilon) n_s^*(\varepsilon) = g_s - \Gamma_s^*(\varepsilon) n_s^*(\varepsilon) + B_s^*(\varepsilon). \quad (2.3)$$

These include the broadenings of the levels of the centers

$$\Gamma_{i\pm}(\varepsilon) = \frac{1}{\tau_1} + \frac{1}{\rho_0} \int \frac{d^3q}{(2\pi)^3} \int d\omega \Delta(\omega, q) n(\omega, q) \times \left\{ \frac{1}{\tau'} \Delta_{i\pm}(\varepsilon + \omega) + \frac{1}{\tau''} \Delta_{i\mp}(\varepsilon + \omega) \right\}, \quad (2.4)$$

$$\Gamma_{i\pm}(\varepsilon) = \frac{1}{\tau_2} + \frac{1}{\rho_0} \int \frac{d^3q}{(2\pi)^3} \int d\omega \Delta(\omega, q) [n(\omega, q) + 1] \times \left\{ \frac{1}{\tau'} \Delta_{i\pm}(\varepsilon - \omega) + \frac{1}{\tau''} \Delta_{i\mp}(\varepsilon - \omega) \right\}. \quad (2.5)$$

Here τ' is the lifetime of level 2 relative to the transition to level 1 in spontaneous emission of a phonon without spin flip, i.e., the time of the transitions $2^\pm \rightarrow 1^\pm$; analogously, τ'' is the time of the $2^\pm \rightarrow 1^\mp$ transitions with spin flip (see Fig. 1). The total lifetime τ of the level 2 relative to spontaneous emission of the phonon is determined by the relation $\tau^{-1} = \tau'^{-1} + \tau''^{-1}$. The times τ_1 and τ_2 have the same meaning as in Sec. 1.

Next, the arrival terms in (2.3) are given by

$$B_{i\pm}(\varepsilon) = \frac{1}{\rho_0} \int \frac{d^3q}{(2\pi)^3} \int d\omega \Delta(\omega, q) [n(\omega, q) + 1] \times \left\{ \frac{1}{\tau'} \Delta_{i\pm}(\varepsilon + \omega) n_{i\pm}(\varepsilon + \omega) + \frac{1}{\tau''} \Delta_{i\mp}(\varepsilon + \omega) n_{i\mp}(\varepsilon + \omega) \right\}, \quad (2.6)$$

$$B_{i\pm}(\varepsilon) = \frac{1}{\rho_0} \int \frac{d^3q}{(2\pi)^3} \int d\omega \Delta(\omega, q) n(\omega, q) \times \left\{ \frac{1}{\tau'} \Delta_{i\pm}(\varepsilon - \omega) n_{i\pm}(\varepsilon - \omega) + \frac{1}{\tau''} \Delta_{i\mp}(\varepsilon - \omega) n_{i\mp}(\varepsilon - \omega) \right\}. \quad (2.7)$$

The terms g_s in (2.3) describe the level population due to the optical pumping. The equations for the centers do not indicate explicitly the parametric variables v , H and r , t that are contained in them and on which n_s^\pm , Γ_s^\pm , and Δ_s^\pm depend. The variables v and H enter as parameters because the phonon-mediated transitions take place within one center. The pumps g_s are assumed to be the same for all types of centers and for both components of the doublet. The variables r and t enter in the right-hand side as parameters for another reason: assuming the inhomogeneity and the nonstationarity to be small, we calculated the interaction of the centers with the phonons in the same manner as in a homogeneous and stationary system.

The phonon balance equation is

$$\Delta(\omega, q)^{-1} \left(\frac{\partial}{\partial t} + v_q \nabla \right) \Delta(\omega, q) n(\omega, q) = S(\omega, q) - \gamma^*(\omega, q) n(\omega, q) + B(\omega, q). \quad (2.8)$$

It includes the phonon width due to the interaction with the centers:

$$\gamma^*(\omega, q) = \frac{N}{\rho_0 \tau'} \text{Av} \int d\varepsilon \Delta_{i+}(\varepsilon + \omega) \Delta_{i+}(\varepsilon) [n_{i+}(\varepsilon) - n_{i+}(\varepsilon + \omega)] + \dots + \frac{N}{\rho_0 \tau''} \text{Av} \int d\varepsilon \Delta_{i+}(\varepsilon + \omega) \Delta_{i-}(\varepsilon) [n_{i-}(\varepsilon) - n_{i+}(\varepsilon + \omega)] + \dots \quad (2.9)$$

The dots in the last formula stand for terms obtained from those written out by replacing (+) by (-) and vice versa. The symbols "Av" denotes averaging over the centers of all types, i.e. over v and H ; the total concentration of the centers is N . The arrival term due to the interaction of the phonons with the centers is of the form

$$B(\omega, q) = \frac{N}{\rho_0 \tau'} \text{Av} \int d\varepsilon \Delta_{i+}(\varepsilon + \omega) \Delta_{i+}(\varepsilon) n_{i+}(\varepsilon + \omega) + \dots + \frac{N}{\rho_0 \tau''} \text{Av} \int d\varepsilon \Delta_{i+}(\varepsilon + \omega) \Delta_{i-}(\varepsilon) n_{i+}(\varepsilon + \omega) + \dots \quad (2.10)$$

It is seen from the last two formulas that γ^* and B are

in fact independent of q . The physical reason for this is the random disposition of the centers. The term $S(\omega, q)$ is responsible for the phonon-phonon interactions.

We have written out above only the broadenings Γ_s^\pm and γ^* ; the corresponding level shifts ΔE_s^\pm and $\Delta\omega$ can be easily obtained from (2.4), (2.5), and (2.9) with the aid of the dispersion relations.

We discuss now the assumptions made in the derivation of the equations for the centers and the phonons. The assumption that the excitations of various centers are not correlated and that the excitation of each center is not coherent can be justified, of course, only for a definite form of optical pumping. Optical pumping in ruby is realized in nonradiative transitions to the levels 1 and 2 from the broad absorption band which lies above them (Fig. 1). The details of the nonradiative transitions are unknown, but it is most likely that if the pumping to the absorption band does introduce some degree of coherence into the system, this coherence is lost in the case of a multiphonon nonradiative transition. (The energy of the ${}^4F_2 \rightarrow {}^2E$ transition in ruby is 3500 cm^{-1} .) The same circumstance justifies the assumption that g_s is the same for both components of the doublet.

The correlations that are established in the system independently of the degree of correlation of the excitation are determined by the "width of the band of occupied states," i.e., in our case by the total width $\delta\omega$ of the $2 \rightarrow 1$ transition, which consists of the inhomogeneous with $\Delta\omega_0$ and the "homogenous broadening" τ^{-1} . (The width $\delta\omega$ is the analog of the thermal width T for equilibrium systems with continuous spectrum.) The assumption that the system is weakly nonstationary is equivalent to assuming that the duration of the investigated processes exceeds the correlation time $\delta\omega^{-1}$, and the assumption that the inhomogeneity is weak is tantamount to assuming that the system dimensions exceed the correlation length $v\delta\omega^{-1}$, where v is the speed of sound.

Let us estimate these quantities for ruby. There are no direct data for $\Delta\omega_0$; judging from the displacements of the lines R_1 and R_2 upon deformation,^[19] $\Delta\omega_0$ should be approximately 20% of the width of these lines, which in turn is $\approx 0.1 \text{ cm}^{-1}$; in other words, $\Delta\omega_0 \approx 0.02 \text{ cm}^{-1}$, which corresponds to a time 0.25 nsec. The time τ is not known exactly; all that is known reliably is that $\tau' + \tau'' = 15 \text{ nsec}$, a value determined from the Orbach data.^[20] As to τ , calculation yields $\tau \approx 1 \text{ nsec}$,^[21] while photon-echo experiments yield $\tau \approx 4 \text{ nsec}$.^[22] In any case it is seen from this that the correlation time in the system is of the order of a fraction of a nanosecond, and the correlation length of the order of hundredths of a millimeter (the average speed of sound is $v = 7.1 \times 10^5 \text{ cm/sec}$, Ref. 23). Yet the time scales of the experiments range from several nanoseconds up to some microseconds, and the spatial scales are not less than a millimeter.

3. WEAK DEPLETION OF THE LOWER LEVEL

In all the experiments with nonequilibrium phonons in ruby, the number of nonequilibrium phonons is small and almost all the metastable centers are in the lower state. In this case the equations become much

simpler.

At $n \ll 1$ it follows directly from (2.5) that $\Gamma_2^* = \tau_2^{-1} + \tau^{-1}$ and $\Delta E_2^* = 0$. The radiative broadening of τ_2^{-1} can be neglected, since $\tau_2 \approx \tau_1 \equiv \tau_R = 4$ msec.^[24] Therefore, reckoning the energy from $E_1 = 0$, we have

$$\Delta_{12}^*(\nu, H; \varepsilon) = \delta(\tau^{-1} | \varepsilon - \omega_0 - \nu \mp \delta_1), \quad (3.1)$$

where

$$\delta(\tau^{-1} | \omega) = \frac{\tau^{-1}}{2\pi} \left[\omega^2 + \left(\frac{1}{2} \tau^{-1} \right)^2 \right]^{-1}. \quad (3.2)$$

It is seen from (2.4) that Γ_1^* consists of two small terms: one τ_R^{-1} and the other proportional to n . We can therefore attempt to seek a solution in which $\Gamma_1^* = 0$ and $\Delta E_1^* = 0$, i.e.,

$$\Delta_{12}^*(\nu, H; \varepsilon) = \delta(\varepsilon \mp \delta_1). \quad (3.3)$$

In addition, we shall assume that $n_1^* \gg n_2^*$.

Substituting (3.3) in (2.7) and (3.9) we can see that we now have only the values $n_1^*(\nu, H; \varepsilon)$ at $\varepsilon = \pm \delta_1$, i.e., the true occupation numbers $n_1^*(\nu, H)$. Since optical excitation is assumed to be nonselective in spin and does not depend on ν or H , and the influence of the phonons on the depletion of the level 1 can be neglected, it is clear that $n_1^*(\nu, H) = n_1$, i.e., it is independent of the spin orientation and of the type of center. We can write for n_1 the obvious balance equation

$$\frac{\partial}{\partial t} n_1 + \frac{1}{\tau_R} n_1 = g_1 + g_2 = g, \quad (3.4)$$

which, of course, follows under the assumptions made also from the equations.

We simplify first the expression for $\gamma^*(\omega)$. We introduce for this purpose the form factor of the transition between unsplit doublets

$$\varphi(\omega) = \delta\omega \int d\nu \varphi(\nu) \delta(\tau^{-1} | \omega - \omega_0 - \nu), \quad (3.5)$$

where $\delta\omega$ is defined by the condition $\varphi(\omega_0) = 1$. Then the H -dependent form factors of the transitions between individual components of the split doublets are

$$\varphi^{\pm\pm}(\omega) = \varphi(\omega \pm \delta_1 \mp \delta_2). \quad (3.6)$$

Substituting (3.1) and (3.3) in (2.9) we get

$$\gamma^*(\omega) = \frac{1}{\tau^*} \Phi(\omega), \quad (3.7)$$

where

$$\frac{1}{\tau^*} = \frac{N^*}{\rho_0 \delta\omega} \frac{1}{\tau}, \quad N^* = 2Nn_1 \quad (3.8)$$

and

$$\Phi(\omega) = A_V \frac{1}{2} \left\{ \zeta' [\varphi^{++}(\omega) + \varphi^{--}(\omega)] + \zeta'' [\varphi^{+-}(\omega) + \varphi^{-+}(\omega)] \right\}, \quad (3.9)$$

$$\zeta' = \tau/\tau', \quad \zeta'' = \tau/\tau''.$$

The meaning of the time τ^* is clearest at $H=0$, when $\Phi(\omega) = \varphi(\omega)$. Omitting factors of the order of unity, we can write

$$\frac{1}{\tau^*} \approx N^* \left(\frac{\tau^{-1}}{\delta\omega} \right) \lambda_0^2 v, \quad (3.10)$$

where λ_0 is the wavelength of the resonant phonon. It suffices now to note that N^* is the concentration of the metastable centers, the parentheses contain the fraction of the centers that are resonant for the given phonon,

and λ_0^2 is the resonant-scattering cross section. Thus, τ^* is the lifetime of the phonon relative to resonant absorption (at the line center).

We proceed now to the transformation $B(\omega)$. Substituting (3.3) in (2.10) we get

$$B(\omega) = \frac{N}{\rho_0 \tau} A_V \{ \Delta_{12}^*(\omega + \delta_1) n_2^*(\omega + \delta_1) + \dots \} + \frac{N}{\rho_0 \tau''} A_V \{ \Delta_{12}^*(\omega - \delta_1) n_2^*(\omega - \delta_1) + \dots \}. \quad (3.11)$$

The dots stand here for terms in which (+) is replaced by (-) and the signs of δ have been reversed. For n_2^* , in turn, we get from (2.3), the equation

$$\left[\frac{\partial}{\partial t} + \frac{1}{\tau} \right] n_2^*(\varepsilon) = g_2 + \frac{n_1}{\rho_0} \left\{ \frac{1}{\tau'} s(\varepsilon \mp \delta_1) + \frac{1}{\tau''} s(\varepsilon \pm \delta_1) \right\}, \quad (3.12)$$

where

$$s(\omega) = \int \frac{d^3 q}{(2\pi)^3} s(\omega, q), \quad s(\omega, q) = \Delta(\omega, q) n(\omega, q). \quad (3.13)$$

Since the true phonon occupation number is

$$n_q = \int d\omega s(\omega, q), \quad (3.14)$$

it is clear that $s(\omega, q)\omega_q$ gives the phonon-field spectral energy density stored in the mode q , while $\omega_0 s(\omega)$ gives the total spectral density of the resonant phonon field.

The solution $n_2^*(\varepsilon)$ of Eq. (3.12) can be represented as a sum of two terms: one proportional to the optical pumping g_2 , and the other proportional to the phonon spectral density $s(\omega)$. The former is equal simply to $\tau \bar{g}_2$, where

$$\bar{g}_2(t) = \int_{-\infty}^t \frac{dt'}{\tau} \exp\left(-\frac{t-t'}{\tau}\right) g_2(t'), \quad (3.15)$$

and the second can be expressed in terms of a function $F(\omega, t)$ that satisfies the equation

$$\left[\frac{\partial}{\partial t} + \frac{1}{\tau} \right] F(\omega) = \frac{1}{\tau} s(\omega), \quad F(\omega, -\infty) = 0. \quad (3.16)$$

In the experiment one usually measures the total number of the metastable centers on the upper level, namely

$$N_2 = N A_V \left\{ \int d\varepsilon n_2^*(\varepsilon) \Delta_{12}^*(\varepsilon) + \dots \right\}. \quad (3.17)$$

We introduce the total number of metastable centers produced by the optical pumping in 1 cm^3 per second, i.e., $G = 2N_g$, as well as the fraction η of the centers excited on the upper level, i.e., $\eta = g_2/g_1$. Then, using (3.1), (3.12), and (3.16), we can transform (3.17) into

$$N_2 = \eta \bar{G} \tau + \int d\omega F(\omega) \Phi(\omega), \quad (3.18)$$

where \bar{G} is calculated from G in analogy with (3.15). This relation reveals simultaneously the meaning of the function $F(\omega)$: the spectral density of the excitation energy of the centers, due to the presence of phonons, is $\omega_0 F(\omega) \Phi(\omega)$.

If we substitute in (2.1) the quantity $n_2^*(\varepsilon)$ in the form of a sum of two terms, then $B(\omega)$ will also break up into two terms. The term proportional to the optical pumping is easy to calculate; it is equal to

$$(\eta \bar{G}/\rho_0 \delta\omega) \Phi(\omega). \quad (3.19)$$

The second term, proportional to $s(\omega)$, is expressed in terms of $F(\omega)$. We leave out the prolonged but rather elementary manipulations and present the final equation

for $s(\omega, \mathbf{q})$, namely

$$\left(\frac{\partial}{\partial t} + \mathbf{v}_e \nabla \right) s(\omega, \mathbf{q}) = S(\omega, \mathbf{q}) - \gamma^*(\omega) s(\omega, \mathbf{q}) + B^*(\omega, \mathbf{q}) + \frac{\eta G}{\rho_0 \delta \omega} \Delta(\omega, \mathbf{q}) \Phi(\omega). \quad (3.20)$$

The term responsible for the phonon anharmonicity is

$$S(\omega, \mathbf{q}) = \Delta(\omega, \mathbf{q}) S(\omega, \mathbf{q}). \quad (3.21)$$

The terms responsible for the reradiation of the phonons are $\gamma^* s$ and B^* , where the corresponding arrival term is

$$B^*(\omega, \mathbf{q}) = \frac{\Delta(\omega, \mathbf{q})}{\rho_0} \frac{1}{\tau} \left\{ F(\omega) \Phi(\omega) + \int d\omega' R(\omega, \omega') [F(\omega') - F(\omega)] \right\}. \quad (3.22)$$

The integral kernel in this equation is expressed in the form of a mean value over the magnetic fields:

$$R(\omega, \omega') = R(\omega', \omega) = \zeta' \zeta'' \cdot \frac{1}{2} \text{Av}_{\mathbf{H}} \{ \delta(\omega - \omega' + 2\delta_1) [\varphi^{++}(\omega) + \varphi^{+-}(\omega)] + \delta(\omega - \omega' - 2\delta_1) [\varphi^{-+}(\omega) + \varphi^{--}(\omega)] \}. \quad (3.23)$$

Using (3.16) to express $F(\omega)$ and substituting in (3.22), we obtain ultimately

$$\begin{aligned} B^*(\omega, \mathbf{q}; \mathbf{r}, t) &= \frac{1}{\tau} \frac{\Delta(\omega, \mathbf{q})}{\rho_0} \left\{ \bar{s}(\omega; \mathbf{r}, t) \Phi(\omega) \right. \\ &\quad \left. + \int d\omega' R(\omega, \omega') [\bar{s}(\omega'; \mathbf{r}, t) - \bar{s}(\omega; \mathbf{r}, t)] \right\}, \\ \bar{s}(\omega; \mathbf{r}, t) &= \int_{-\infty}^t \frac{dt'}{\tau} \exp \left[-\frac{t-t'}{\tau} \right] s(\omega; \mathbf{r}, t'). \end{aligned} \quad (3.24)$$

The term non-integral in the frequencies in (3.24) describes reradiation processes that do not alter the spin state of the metastable center on the level 1 (reradiation without spin flip, see Fig. 2), and the integral term stands for processes in which the spin state of the center on the level 1 changes after the reradiation (reradiation with spin flip). In the first process are coupled only the components of the spectral density $s(\omega, \mathbf{q})$ with a single frequency ω , while in the second are coupled components with frequencies that differ by the splitting of the lower level $2\delta_1$. It is obvious that the splitting of the upper level does not lead to such an effect. An indication that the frequency is changed by reradiation with spin flip is contained in Ref. 6.

Equation (3.4) can be rewritten in the form

$$\left[\frac{\partial}{\partial t} + \frac{1}{\tau_e} \right] N^* = G. \quad (3.25)$$

Its solution yields the distribution of the concentration of the metastable atoms $N^*(\mathbf{r}, t)$. The width $\gamma^*(\omega)$ then becomes, according to (3.8), a known function of \mathbf{r}, t , and ω ; from the dispersion relations we can reconstruct $\Delta\omega(\omega; \mathbf{r}, t)$ and obtain $\Delta(\omega, \mathbf{q}; \mathbf{r}, t)$ from (2.2). It remains now to solve Eq. (3.20) for the function $s(\omega, \mathbf{q}; \mathbf{r}, t)$ with an arrival term in the form (3.24). Calculating next $s(\omega)$, we can find from (3.16) the function $F(\omega; \mathbf{r}, t)$. Knowledge of F makes it possible to calculate from (3.18) the spatial distribution and the time dependence of the R_2 -luminescence, which is proportional to $N_2(\mathbf{r}, t)$.

It is necessary to add to (3.20) the conditions imposed on $s(\omega, \mathbf{q})$ on the crystal boundary; these conditions are determined by the character of the phonon reflection from the helium-crystal interface. The excitation of the system by the thermal momentum also enters via the boundary conditions: besides the reflection conditions, an additional flux $\mathbf{v}_e s(\omega, \mathbf{q})$ is specified on the

crystal-emitter boundary for \mathbf{v}_e directed in the interior of the crystal.

In conclusion, we discuss the criterion for the validity of the approximations used to simplify the equations. The widths of all the functions in terms of ω and ω_e are determined by the quantities τ^{*-1} and $\delta\omega$. Therefore the approximation $\Gamma_1 = 0$ means in fact that $\Gamma_1 \ll \delta\omega$ and $\Gamma_1 \ll \tau^{*-1}$. On the other hand it is seen from (2.4) that $\Gamma_1 \approx \tau_R^{*-1} + n\tau^{-1}$. Since $\delta\omega \gtrsim \tau^{-1} \gg \tau_R^{*-1}$, the condition on Γ_1 reduces at $n \ll 1$ to the inequality $n\tau^{-1} \ll \tau^{*-1}$ or, equivalently, $n \ll \alpha$, where $\alpha \equiv N^*/\rho_0 \delta\omega$. This parameter has a simple meaning: it is the ratio of the number of electronic degrees of freedom to the number of phonon degrees of freedom in the band $\delta\omega$; in the theory of spin-lattice relaxation it is sometimes called the narrowness factor of the phonon bottleneck. Thus, the condition for the applicability of the equations of the present section is

$$n \ll 1, \alpha; \quad N_* \ll N.$$

By way of example, we mention that in ruby $\rho_0 = 1.3 \times 10^7 \text{ sec} \cdot \text{cm}^{-3}$ (for the three phonon branches), so that a typical value is $\rho_0 \delta\omega \approx 10^{17} \text{ cm}^{-3}$. Typical values of N^* range from 10^{14} to 10^{18} cm^{-3} .

4. SPECTRAL DIFFUSION

If there is no external magnetic field, then the splitting $2\delta_1$ is connected only with local magnetic fields. The splitting can be estimated in this case from the width of the EPR line on the metastable level 1. In ruby at $N = 2 \times 10^{19} \text{ cm}^{-3}$ (0.05% of Cr^{3+} by weight) we have $2\delta_1 = 1.5 \times 10^{-3} \text{ cm}^{-1}$ according to Ref. 20. Since $2\delta_1 \ll \delta\omega$, the change of the frequency in reradiation with spin flip can be visualized as spectral diffusion. Expanding, as usual, in terms of δ_1 and δ_2 , we can easily transform the integral term of (3.24) into a differential term:

$$\frac{1}{\tau} \int d\omega' R(\omega, \omega') [s(\omega') - s(\omega)] = \frac{\partial}{\partial \omega} \left[D_s(\omega) \frac{\partial}{\partial \omega} s(\omega) \right]. \quad (4.1)$$

We have introduced here the spectral-diffusion coefficient

$$D_s(\omega) = \zeta \frac{\delta^2}{\tau} \varphi(\omega), \quad (4.2)$$

where

$$\delta^2 = \text{Av}_{\mathbf{H}} \delta_1^2, \quad \zeta = 4\zeta' \zeta'' = 4\tau / (\tau' + \tau'') < 1. \quad (4.3)$$

The spectral diffusion is significant at times t such that

$$(\zeta \delta^2 / \tau') t \gtrsim (\delta\omega)^2. \quad (4.4)$$

The value of ζ in ruby, as seen from the data at the end of Sec. 2, lies in the interval from ~ 0.1 to ~ 1 . A typical time of the long-range R_2 -luminescence decay, observed in the experiment, is $\tau_{R_2} \approx 1 \mu\text{sec}$. Therefore the spectral diffusion is significant at $N^* \gtrsim 10^{16} - 10^{17} \text{ cm}^{-3}$. Experiments on the initial decay of R_2 luminescence yielded^[17, 25] $\tau_{R_2} \approx 10 \text{ nsec}$; in this case the spectral diffusion is apparently inessential.

Spectral diffusion should be distinguished from redistribution over the frequencies in the Holstein-Bberman equations, although qualitatively it can sometimes

lead to analogous effects. First, spectral diffusion calls for splitting of the lower level; second, it is characterized by a definite rate and is not instantaneous as in the Holstein-Biberman equations.

Turning on an external magnetic field H_0 enhances the spectral diffusion. If the field H_0 is weak, so that the splitting $\delta_1(H_0)$ it produces is small compared with $\delta\omega$, then this field can be taken into account by changing δ^2 in the diffusion coefficient. On the other hand if the field is strong and $\delta_1(H_0) \gtrsim \delta\omega$, then the redistribution over the frequency spectrum does not have the character of diffusion. It is seen from (3.4) that the role of the magnetic field does not reduce in this case to a "dragging asunder" of the phonon packets, as is sometimes tacitly assumed—the redistribution among them must also be taken into account.

5. COMPARISON WITH THE BIBERMAN-HOLSTEIN EQUATIONS

If there is no external magnetic field, and the local fields can be neglected, then the equation for $s(\omega, q)$ takes the following form:

$$\left(\frac{\partial}{\partial t} + v_q \nabla \right) s(\omega, q) = -\gamma^*(\omega) s(\omega, q) + \frac{\Delta(\omega, q)}{\rho_0} \frac{1}{\tau} \varphi(\omega) F(\omega) + S(\omega, q) + \frac{\eta G}{\rho_0 \delta\omega} \Delta(\omega, q) \varphi(\omega). \quad (5.1)$$

Let us compare this equation with (1.14) and Eq. (3.16) for $F(\omega)$ with (1.15). It can be stated that the crossing terms of the system (3.16), (5.1) describe reradiation of spectral density $s(\omega, q)$ with conservation of ω and with variation of q (as should be the case with scattering by an immobile center); after the reradiation, the spectral density at a given ω is distributed over q in accord with $\Delta(\omega, q)$. It is important, however, that the probability of the reradiation depends not on q but on ω ; therefore after the reradiation this probability does not change. This is the essential difference between our equations and the Holstein-Biberman equations.

It is instructive also to understand when Eq. (5.1) reduces to the ordinary kinetic equation for n_q , in other words, when we can confine ourselves to consideration of $s(\omega, q)$ on the mass shell $\omega = \omega_q$. It is seen from (5.1) that for this purpose it is necessary to be able to make the replacement $\Delta(\omega, q) \rightarrow \delta(\omega - \omega_q)$. Only then can we seek a solution in the form $s(\omega, q) = n_q \delta(\omega - \omega_q)$ and, cancelling out $\delta(\omega - \omega_q)$, we obtain an equation for n_q . This procedure is obviously possible only when the width of the function of $\Delta(\omega, q)$ in terms of ω is less than the width of the function $\varphi(\omega)$, i.e., when $\tau^{*-1} \ll \delta\omega$.

6. SPATIAL DIFFUSION

It is known that the Biberman-Holstein equations do not go over into the diffusion equation for the total phonon concentration $n(r, t)$. The reason is that the reradiation process increases the number of phonons with larger mean free paths relative to resonant absorption. In our Eq. (5.1), as already indicated, the mean free path relative to absorption by the centers is not charged in the course of reradiation of the spectral density. Therefore in the case when the nonstationarity and spatial inhomogeneity are small not only compared with

$\delta\omega$ but also compared with τ^{*-1} , i.e., when the reradiation proceeds rapidly enough, Eq. (5.1) can be reduced to the diffusion equation for the total spectral density $s(\omega, r, t)$.

Naturally, the spatial diffusion can describe only the resonant components of the spectral density, i.e., $s(\omega)$ with ω close to ω_0 —the other components of $s(\omega)$ are infrequently reabsorbed. For the components with $\omega \approx \omega_0$ we can approximate

$$S(\omega, q) = -\tau_0^{-1} [s(\omega, q) - s_T(\omega, q)] \quad (6.1)$$

with a relaxation time τ_0 that does not depend on ω and q . Here s_T is the equilibrium spectral density, whose value at ω close to ω_0 is

$$s_T(\omega, q) = n_0 \delta(\omega - \omega_0), \quad n_0 = \exp(-\omega_0/T), \quad T \ll \omega_0, \quad (6.2)$$

(for ruby $\omega_0 = 29 \text{ cm}^{-1} = 42 \text{ K}$).

To obtain the diffusion equation, we resolve, as usual $s(\omega, q)$ into two components s^+ and s^- , even and odd in q . Subdividing correspondingly Eq. (5.1) and retaining only the principal terms, we get

$$\frac{1}{\tau} s^+(\omega, q) = \frac{\Delta(\omega, q)}{\rho_0} \frac{1}{\tau} F(\omega), \quad (6.3)$$

$$v_q \nabla s^+(\omega, q) + \gamma^*(\omega) s^-(\omega, q) = 0. \quad (6.4)$$

It follows from (6.4) that $s^- \ll s^+$. From (6.3) we have

$$s(\omega, q) = \frac{\Delta(\omega, q)}{\rho_0} s(\omega), \quad F(\omega) = \frac{\tau}{\tau_0} s(\omega). \quad (6.5)$$

We integrate (5.1) with respect to q and add to Eq. (3.16) multiplied by $\varphi(\omega)$. We then obtain the law of conservation of the spectral density in the phonons + centers system:

$$\begin{aligned} \frac{\partial}{\partial t} [s(\omega) + \varphi(\omega) F(\omega)] + \nabla \int \frac{d^3 q}{(2\pi)^3} v_q s(\omega, q) \\ = -\frac{1}{\tau_0} [s(\omega) - n_0 \rho_0] + \eta \frac{G}{\delta\omega} \varphi(\omega). \end{aligned} \quad (6.6)$$

We substitute here in place of $F(\omega)$ its expression in terms of $s(\omega)$ from (6.5); it is possible to replace s by s^- under the integral sign, and express s^- in terms $\nabla s^+ \approx \nabla s$ from (6.4). We then obtain the diffusion equation of interest to us:

$$\begin{aligned} \frac{\partial}{\partial t} \{s(\omega) [1 + \alpha \varphi(\omega)]\} - \operatorname{div} \{D_v(\omega) \operatorname{grad} s(\omega)\} \\ = -\frac{1}{\tau_0} [s(\omega) - n_0 \rho_0] + \eta \frac{G}{\delta\omega} \varphi(\omega), \end{aligned} \quad (6.7)$$

where we have introduced the coefficient of spatial diffusion of the spectral density

$$D_v(\omega) = \tau_0 v^2 \tau \varphi(\omega)^{-1}. \quad (6.8)$$

The presence of an additional term proportional to $\alpha \varphi(\omega)$ under the sign of the time derivative is connected with the fact that part of the energy in the system has been accumulated in the form of excitation of the centers. This part, however, does not contribute to the energy transfer; there is therefore no such term in the flux.

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Screening of charges and Friedel oscillations of the electron density in metals having differently shaped Fermi surfaces

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Analytic and numerical integration methods are used to obtain the spatial distribution of the screened Coulomb potential of point charges in the interior and on the surface of metals having different Fermi-surface shapes. It is shown that in isotropic metals with quasi-two-dimensional or quasi-one-dimensional electron spectra the Friedel oscillations of the electron density decreases along normals to a cylindrical or plane Fermi surface, like $r^{-2} \sin 2k_F r$ or $r^{-1} \cos 2k_F r$, respectively, and attenuate exponentially in perpendicular directions. Along the surface of an isotropic metal with a spherical Fermi surface, the Friedel oscillations decrease like $r^{-5/2} \cos 2k_F r$. In the Thomas–Fermi approximation, the screened charge potential in a homogeneous metal takes the form $e^{-\alpha r}/r$ regardless of the shape of the Fermi surface, and along the surface of a semi-infinite metal it decreases in power-law fashion like r^{-3} . An expression is obtained for the potential energy of the charges near the surface of a metal. This expression, together with the image forces, describes the Friedel oscillations and the dipole-dipole interactions. The results explain, in particular, the experimental data on the ordering of adsorbed Sr and La films on the (112) faces of W and Mo single crystals.

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1. INTRODUCTION

It is known that the steplike character of the Fermi distribution of the conduction electrons in metals leads to the appearance of the so-called Friedel oscillations of the screened Coulomb potential, which by virtue of the spherical Fermi surface over large distances from the charge, $r \gg k_F^{-1}$ (k_F is the Fermi momentum, $\hbar = 1$) decrease like $r^{-3} \cos 2k_F r$.^[1,2] For the same reason, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction between the magnetic moments of nuclei or paramagnetic impurities in an isotropic metal behaves asymptotically like r^{-3} .

The law governing the decrease of the Friedel oscillations depends, however, on the “dimensionality of the metal” d , i.e., on whether we are dealing with a bulky metal, with a film, or with a thin filament. Therefore even in the case of a three-dimensional isotropic electron spectrum, as shown by Adawi,^[4] the perturbation δn of the electron density decreases like $r^{-5/2} \cos 2k_F r$ at $d=2$ and like $r^{-2} \sin 2k_F r$ at $d=1$. If at the same time the electron spectrum is two-dimensional at $d=2$ and one-dimensional at $d=1$, then the screened Coulomb potential decreases respectively like $r^{-2} \sin 2k_F r$ (Ref. 5) and $r^{-1} \cos 2k_F r$.^[6]