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ROLE OF LATTICE THERMAL CONDUCTIVITY IN THE PHENOMENOLOGICAL THEORY OF PARAMAGNETIC RELAXATION

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The thermal conductivity of the lattice is introduced into the phenomenological theory of paramagnetic relaxation[1] in parallel fields. It is assumed that the surface of the spherical sample is isothermal. Lattice thermal conductivity is found to be important at low temperatures (helium temperatures and below).

1. The first phenomenological theory of paramagnetic relaxation in parallel fields, developed by Casimir and Du Pré,[2] in which only spin-lattice relaxation was considered, was extended by Eisenstein[3] to take account of lattice relaxation due to the finite thermal conductivity of the lattice; it was assumed that the surface of the paramagnetic sample is isothermal. In the present work we consider lattice relaxation in conjunction with spin-lattice relaxation and spin relaxation. The spin relaxation is considered only within the framework of the Shaposhnikov analysis,[1] i.e., spin relaxation is considered without taking account of the modifications which have been introduced recently in Ref. 4. Thus, we are extending the Shaposhnikov analysis in the same way as Eisenstein extended the Casimir-Du Pré theory. This extension and the evaluation of the roles played by all three of the above-mentioned relaxation processes at various field strengths, frequencies, and temperatures are of interest for two reasons: on the one hand, at helium temperatures and low frequencies, where the Casimir-Du Pré theory does not agree with experiment, relaxation within the lattice is important (cf. Ref. 3); on the other hand, at high temperatures and frequencies the spin relaxation becomes important, assuming a decisive influence at room temperatures and ultrahigh frequencies (cf. Refs. 5 — 7).

2. Before turning to the analysis indicated above, we shall consider the question of whether or not a complete theory of paramagnetic relaxation should take account of still one other relaxation process — relaxation within the spin system, due to the finite thermal conductivity of this system.

The relaxation times θ_l and θ_s, which characterize the rate of dissipation of temperature inhomogeneities in the lattice and in the spin system, are proportional respectively to the ratios c_l/λ and c/λ_s where c_l is the heat capacity of the lattice, c is the heat capacity of the spin system, and λ and λ_s are the thermal conductivities of the lattice and spin system. Thus the ratio of relaxation times θ_s and θ_l is given by:

$$\theta_s / \theta_l \sim c\lambda / c_l\lambda_s. \tag{1}$$

To make a rough estimate it is sufficient to consider the behavior of $\theta_s\omega$ at frequencies for which $\theta_l\omega \sim 1$; in the indicated frequency region, from the relation in (1) we have

$$\theta_s\omega \sim c\lambda / c_l\lambda_s. \tag{2}$$

Information as to c, c_l, and λ for various materials can be determined over a rather wide temperature range. As far as λ_s, however, we are familiar only with the work of Akhiezer and Pomeranchuk,[8] who obtained results for thermal conductivity of paramagnetic materials at very low temperatures; at these temperatures the small-excitation method can be used. At temperatures of 0.14 — 0.3° K and for $H = 0$, λ_s in potassium chrome alum was found to be approximately 5×10^3 times smaller than λ.[8,9] At these same temperatures and in the absence of a magnetic field it is found that $c/c_l > 10^3$ (Ref. 10). In the frequency region in which the relation in (2) applies, we have $\theta_s\omega > 10^6$; consequently, the thermal conductivity of the spin system is not important in the indicated temperature region in the absence of an external magnetic field at frequencies for which lattice relaxation is of importance. These frequencies are approximately 10^4 sec^{-1} because for the temperatures being considered $\theta_l \sim 10^{-4}$ sec, as is shown by estimates carried out for a number of salts of elements of the iron group (cf. below, Sec. 7). However, there is a factor of 10^6 in the right-hand part of the inequality given above. Hence our conclusion that the thermal conductivity of the spin system can be neglected holds for the entire frequency region used in studying paramagnetic relaxation. Obviously this conclusion is all the more justified when an external magnetic field is applied or at higher temperatures since c_{ℓ} and λ do not change as the field is increased, c does not change or changes slowly, and λ_S falls off very rapidly as the temperature is increased. The rapid decrease in λ_S predominates over the variation of the other quantities which appear in (2). It is therefore reasonable to assume that the thermal conducitivity of the spin system is not of any great importance in paramagnetic relaxation (it should be noted, however, that the data on λ_S used in the present estimates refer only to the very low temperatures considered in Ref. 8).

3. Following Eisenstein,[3] we consider the problem of a spherical paramagnetic sample at the surface of which a constant temperature is maintained. The choice of a spherical geometry is occasioned by the fact that of all the simple shapes the sphere is the one which most closely approximates the shape of the samples used in experiments (cf. Ref. 11, for example). As far as the constant temperature at the surface of the sample is concerned it must be noted that an analysis of the degree to which this situation corresponds to the experimental conditions is not a simple one and requires individual attention in each different experimental case. There is always, however, some exchange of heat between the surface of the sample and the medium which surrounds it. Hence, in any case, it may be assumed that an analysis of paramagnetic relaxation under the assumption of isothermal conditions at the surface of the sample, such as used by Eisenstein,[3] is closer to reality than earlier analyses in which the temperature of the lattice (hence, the spin system) was assumed to be uniform over the entire sample.

We consider a solid non-conducting paramagnetic material with pure spin magnetism which is magnetically isotropic. The paramagnetic material is placed in an external magnetic field of fixed direction and magnitude which depends on the time t as follows:

$$H(t<0)=H_0, \quad H(t\geqslant 0)=H_0+\eta_0 e^{i\omega t}. \tag{3}$$

4. In a phenomenological theory of paramagnetic relaxation which treats both the spin-lattice relaxation and the spin relaxation attention must be paid to the fact that when the paramagnetic material is placed in an alternating magnetic field its spin system passes through non-equilibrium states (cf. Refs. 1 and 4). Applying non-equilibrium thermodynamics (the justification for applying this analysis in paramagnetic relaxation is discussed in Refs. 1 and 12) in the general scheme used earlier[1,4] and taking account of the thermal conductivity of the lattice the following equations are obtained:

$$\Phi_{TM}\Phi_{MM}\tau_l\dot{\xi}+\Phi_{TT}\Phi_{MM}\tau_l(\dot{\vartheta}+\dot{\vartheta}_l)$$

$$-(\Phi_{TM}^2-\Phi_{TT}\Phi_{MM})\vartheta=\Phi_{TH}\Phi_{MM}\tau_l\dot{\eta}, \tag{4}$$

$$\Phi_{MM}\tau_s\dot{\xi}+\Phi_{MM}\xi+\Phi_{TM}(\vartheta+\vartheta_l)=-\Phi_{HM}\eta, \tag{5}$$

$$c_l\dot{\vartheta}_l=\lambda\Delta\vartheta_l+\alpha\vartheta, \tag{6}$$

where

$$\tau_l\equiv T_0\alpha^{-1}\Phi_{MM}^{-1}(\Phi_{TM}^2-\Phi_{TT}\Phi_{MM}), \tag{7}$$

$$\tau_s\equiv \varkappa^{-1}\Phi_{MM}^{-1}. \tag{8}$$

Here ξ is the variable part of the magnetization M, ϑ is the difference between the temperature of the spin system T and the lattice temperature, ϑ_{ℓ} is the difference in the temperature of the lattice and the fixed temperature T_0 at the surface of the sample, the Φ indices are the partial derivatives of the non-equilibrium thermodynamic potential of the spin system $\Phi(H, T, M)$ taken at $H = H_0$, $T = T_0$, and $M = M_0$ where M_0 is the equilibrium value of the magnetization, corresponding to $T = T_0$ and $H = H_0$, $\alpha(H_0, T_0)$ is the coefficient of heat conduction between the spin system and the lattice, $\kappa(H_0, T_0)$ characterizes the rate of change of ξ due to the interaction within the spin system (cf. Refs. 1 and 4), τ_{ℓ} is the spin-lattice relaxation time, which characterizes the rate of equalization of the temperatures of the spin system and the lattice under the assumption that the spin system and the lattice each pass through their own states of internal equilibrium without coming to equilibrium with each other, and T_S is the spin relaxation time, which characterizes the rate at which the magnetization M approaches its equilibrium value M_0 for an isothermal spin system at $T = T_0$ [by virtue of internal interactions in the spin system (cf. Ref. 13)].

It will be assumed that up to $t = 0$, at which time the variable external magnetic field is switched on, the system has been in an equilibrium state corresponding to a magnetic field H_0 and temperature T_0; in this case the initial conditions are that ϑ, ϑ_{ℓ}, and ξ are zero. The boundary conditions are obviously $\vartheta_{\ell} = 0$ at the surface of the sphere and $d\vartheta_{\ell}/dr = 0$ at the center of the sphere. Thus, the initial conditions and boundary conditions for the problem are

$$
\begin{aligned}
&t=0:\quad \vartheta=0,\quad \vartheta_l=0,\quad \xi=0;\\
&r=R:\quad \vartheta_l=0;\quad r=0:\quad (\partial\vartheta_l/\partial r)=0.
\end{aligned}
\tag{9}
$$

5. The system of equations given in (4) — (6) and (9) is solved by means of a Laplace transform. The steady-state solution is used to find the complex magnetic susceptibility as a function of the radial coordinate. In order to obtain the general characteristics of the entire paramagnetic sample this expression must be averaged over the entire sample. The following expression is obtained for the susceptibility χ:

$$\frac{\chi}{\chi_0}=\frac{1+i(1-F)\tau_l\omega+(F^*/G)\left[3\coth(G^{1/2}R)/G^{1/2}R-3/GR^2-1\right]}{1+i(\tau_l+\tau_s)\omega-(1-F)\tau_l\tau_s\omega^2}; \tag{10}$$

$$F^*\equiv iFc_H\lambda^{-1}\omega\left[1-(1-F)\tau_l\tau_s\omega^2+i(\tau_l+\tau_s)\omega\right]^{-1}, \tag{11}$$

$$
\begin{aligned}
G\equiv ic_l\lambda^{-1}\omega\{1+(c_H/c_l)\left[1+i(1-F)\tau_s\omega\right]\left[1-(1-F)\tau_l\tau_s\omega^2\right.\\
\left.+i(\tau_l+\tau_s)\omega\right]^{-1}\},
\end{aligned}
\tag{12}
$$

$$F\equiv 1-\Phi_{TT}\,\Phi_{MM}\left(\Phi_{TT}\,\Phi_{MM}-\Phi_{TM}^2\right)^{-1}, \tag{13}$$

$$\chi_0=-\Phi_{HM}/\Phi_{MM}. \tag{14}$$

The quantity χ_0 is the equilibrium magnetic susceptibility; the quantity F is equal to $1 - c_M/c_H$ where c_M and c_H are the specific heats of the spin system for constant magnetization and for constant field.

6. The general expression given in (10) for χ, the complex magnetic susceptibility, is rather complicated. Hence we consider the two limiting cases in which the argument of the hyperbolic cotangent in (10) is either very small or very large. In the first case

$$\theta_l\omega\ll 1,\quad \theta_l\omega\ll c_l/c_H, \tag{15}$$

where

$$\theta_l=c_lR^2/15\lambda \tag{16}$$

[cf. Eq. (12)] and the following approximate expressions are obtained for the real part χ' and the imaginary part χ'' of the susceptibility

$$
\begin{aligned}
\frac{\chi'}{\chi_0}&=\frac{1+(1-F)\tau_l^2\omega^2}{\left[1-(1-F)\tau_l\tau_s\omega^2\right]^2+(\tau_l+\tau_s)^2\omega^2}\\
&-\frac{2F(\tau_l+\tau_s)\omega\left[1-(1-F)\tau_l\tau_s\omega^2\right]}{\{\left[1-(1-F)\tau_l\tau_s\omega^2\right]^2+(\tau_l+\tau_s)^2\omega^2\}^2}\,\frac{c_H}{c_l}\,\theta_l\omega,
\end{aligned}
\tag{17}
$$

$$
\begin{aligned}
\frac{\chi''}{\chi_0}&=\frac{(F\tau_l+\tau_s)\omega+(1-F)^2\tau_l^2\tau_s\omega^3}{\left[1-(1-F)\tau_l\tau_s\omega^2\right]^2+(\tau_l+\tau_s)^2\omega^2}\\
&+\frac{F\left[1-(\tau_l+\tau_s)^2\omega^2\right]+(1-F)^2\tau_l^2\tau_s^2\omega^4}{\{\left[1-(1-F)\tau_l\tau_s\omega^2\right]^2+(\tau_l+\tau_s)^2\omega^2\}^2}\,\frac{c_H}{c_l}\,\theta_l\omega.
\end{aligned}
\tag{18}
$$

In the second case:

$$\theta_l\omega\gg 1/15\quad\text{or}\quad \theta_l\omega\gg c_l/15\,c_H, \tag{19}$$

and an approximate expression for the complex susceptibility may be written as follows:

$$\frac{\chi}{\chi_0}=\frac{1+i(1-F)\tau_l\omega+(F^*/G)\left[3/\sqrt{G}R-1\right]}{1-(1-F)\tau_l\tau_s\omega^2+i(\tau_l+\tau_s)\omega}. \tag{20}$$

The quantity θ_{ℓ} given in (16) may be considered the lattice relaxation time which determines the rate of dissipation of spatial inhomogeneities in the lattice temperature.

Below we shall examine the approximate expressions given in (17), (18), and (20) to determine the role of lattice relaxation at various temperatures and to investigate the frequency region $\tau_{\ell}\omega \sim 1$ at low (helium) temperatures; the latter is of interest in connection with our analysis of various experiments (cf. below, Sec. 9).

7. To delineate the role of lattice relaxation at various temperatures we consider the temperature dependence of the quantities θ_{ℓ}, τ_{ℓ}, and c_{ℓ}/c_H which determine the temperature variation of the magnetic susceptibility. In a large number of cases the spin-lattice relaxation time, τ_{ℓ}, (for example, almost all investigated compounds of elements of the iron group) at temperatures which are not too low, lies within the limits $10^{-6} - 10^{-8}$ sec;[14] as the temperature is reduced the spin-lattice relaxation time reaches values close to $10^{-2} - 10^{-3}$ sec at helium temperatures.[15,11] The quantity θ_{ℓ} depends on temperature through the heat capacity of the lattice c_{ℓ} and through the coefficient of heat conductivity λ. At temperatures which are appreciably above the Debye temperature of the material c_{ℓ} is temperature independent (cf. Ref. 16 for example). At these temperatures the heat conductivity of dielectric crystals[16] is proportional to T^{-1} so that $\theta_{\ell} \sim T$. At lower temperatures c_{ℓ} diminishes as the temperature is reduced whereas λ exhibits approximately the same temperature dependence ($\lambda \sim T^{-1}$) so that θ_{ℓ} increases with increasing temperature at a still higher rate. At helium temperatures the thermal conductivity falls off rapidly as the temperature is reduced, approximately as T^3, (Refs. 9, 17), while c_ℓ and θ_ℓ remain constant at temperatures below the Debye temperature. In potassium chrome alums in the form of single crystals $\lambda = 5.3 \times 10^5 T^3$ erg-sec^{-1}-deg^{-1} for $T = 0.14 - 0.3$° K,[9] $\rho = 1.8$ g-cm^{-3} (Ref. 3), $c_\ell = 100\ T^3$ erg-g^{-1} (Ref. 10). Whence, from Eq. (16), $\theta_\ell = 2 \times 10^{-5}$ sec; in a powder θ_ℓ becomes longer because of the reduction in λ (Ref. 18). Hence it is clear that for a wide range of temperatures $\theta_\ell/\tau_\ell \gg 1$ and $\theta_\ell/\tau_\ell \sim 1$ and only at helium temperatures does $\theta_\ell/\tau_\ell \ll 1$. Finally, in many salts of elements of the iron group we have $c_\ell/c_H \gg 1$ down to very low temperatures; it is only as the helium temperature is approached that the heat capacity of the lattice becomes comparable with the heat capacity of the spin system. The lattice heat capacity falls off rapidly as the temperature is reduced further. Thus, in a number of salts of the iron group $c_\ell/c_H \sim 10^3$ in a field of 5,000 oersted at 15° K whereas a field of 1,000 oersted is required to obtain this value at 10° K.[10]

We now consider the effect of the lattice heat conductivity on the magnetic susceptibility at various temperatures. We have shown above that a wide temperature region, aside from extremely low temperatures, $\theta_\ell/\tau_\ell > 1$. At all these temperatures, in accordance with our approximation, it is meaningful to consider only the case $\theta_\ell\omega \gg 1/15$ since $\theta_\ell\omega \ll 1$ is realized at frequencies which are small in the sense $\tau_\ell\omega \ll 1$, when the susceptibility is essentially equal to its static value. In the appropriate approximate expression (20) terms proportional to F^*/G and $F^*/G^{3/2}R$ take account of the effect of lattice heat capacity and heat conductivity respectively [cf. Eqs. (11) and (12)]. Because of the ratio F^*/G, these terms are proportional to the quantity c_H/c_ℓ which, as we have seen, is small down to very low temperatures. Moreover, the second of these terms falls off still more rapidly as the temperature increases because of the factor $(G^{1/2}/R)^{-1}$ which, in accordance with Eqs. (12) and (16) is proportional to $(\theta_\ell\omega)^{-1/2}$ (cf. the temperature dependence for θ_ℓ indicated above). Thus we may conclude that the lattice heat conductivity, being considered in the present work, is important only at extremely low temperatures (helium temperatures and below).

8. In accordance with this conclusion, we now consider the helium temperature region; this is the region which is of interest from the point of view of experimental results (cf. Sec. 9). At helium temperatures $\theta_\ell/\tau_\ell \ll 1$, hence in our approximation it is necessary to take the case $\theta_\ell\omega \ll 1$ since when $\theta_\ell\omega \gg 1$ and $\theta_\ell\omega \sim 1$, the condition that $\theta_\ell/\tau_\ell \ll 1$ corresponds to frequencies such that $\tau_\ell\omega \gg 1$; at these frequencies lattice relaxation processes become unimportant. Having in mind a discussion of experiments at helium temperatures, we apply Eqs. (16) and (17) for frequencies such that $\tau_\ell\omega \sim 1$. Inasmuch as $\tau_\ell \sim 10^{-2} - 10^{-3}$ sec at helium temperatures while the spin relaxation time τ_S, as has been shown by many experiments,[14] is temperature independent and of the order of 10^{-9} sec, $\tau_\ell \gg \tau_S$. Hence we may expand Eqs. (17) and (18) in a power series in $\tau_S\omega$ and consider only the linear terms; this procedure yields:

$$\frac{\chi'}{\chi_0} = \left(1 - \frac{F\tau_l^2\omega^2}{1+\tau_l^2\omega^2}\right)\left(1 - \frac{2F\tau_l^2\omega^2}{1+\tau_l^2\omega^2}\frac{\tau_s}{\tau_l} - \frac{2F\tau_l^2\omega^2}{[1+(1-F)\tau_l^2\omega^2](1+\tau_l^2\omega^2)}\frac{c_H}{c_l}\frac{\theta_l}{\tau_l}\right); \tag{21}$$

$$\frac{\chi''}{\chi_0} = \frac{F\tau_l\omega}{1+\tau_l^2\omega^2}\left[1 + \frac{1}{F} + \frac{1+(2(1-F)-F^2)\tau_l^2\omega^2+(1-F)^2\tau_l^4\omega^4}{1+\tau_l^2\omega^2}\frac{\tau_s}{\tau_l} + \frac{1-\tau_l^2\omega^2}{1+\tau_l^2\omega^2}\frac{c_H}{c_l}\frac{\theta_l}{\tau_l}\right]. \tag{22}$$

It is obvious from the expressions obtained that at the frequencies being considered the spin-lattice relaxation, which is governed by the Casimir-Du Pré formula,[2] plays an important role; the second and third terms reflect the effect of spin relaxation and lattice relaxation repsectively. Since the coefficients for τ_S/τ_ℓ and $\theta_\ell c_H/\tau_\ell c_\ell$ are of the order of unity at average fields regardless of temperature, the effect of both of the above-indicated relaxation processes on magnetic susceptibility is determined by the quantities τ_S/τ_ℓ and $\theta_\ell c_H/\tau_\ell c_\ell$. At helium temperatures $\tau_S/\tau_\ell \sim 10^{-7} - 10^{-5}$ while $\theta_\ell c_H/\tau_\ell c_\ell$, as has been indicated by estimates for a number of materials (cf. below, Sec. 9), is of the order of $10^{-2} - 10^{-1}$; whence it is clear that spin relaxation is not important at the temperatures and frequencies being considered. However, at higher temperatures and these same frequencies the spin relaxation process becomes more important whereas the lattice relaxation becomes less important. Thus, at some temperature the effect of both of these processes on magnetic susceptibility should be the same. To determine this temperature and to evaluate the spin relaxation and lattice relaxation as compared with spin-lattice relaxation, we consider the expression in (20). This relation, as has been mentioned above, is of interest at relatively high temperatures for which $c_\ell/c_H \gg 1$ in the frequency region for which $\tau_\ell\omega \sim 1$ (and $\tau_s\omega \ll 1$). Expanding the expression in (20) with $\tau_s\omega \ll 1$ and $c_H/c_\ell \ll 1$ we obtain the following expressions for χ' and χ'':

$$\frac{\chi'}{\chi_0} = \left[1 - \frac{F\tau_l^2\omega^2}{1+\tau_l^2\omega^2} - \frac{1-\tau_l^2\omega^2}{(1+\tau_l^2\omega^2)^2} F\frac{c_H}{c_l}\right]\left[1 - \frac{2F\tau_l^2\omega^2}{1+\tau_l^2\omega^2}\frac{\tau_s}{\tau_l} + \sqrt{0,3}\,\frac{1-\tau_l^2\omega^2-2\tau_l\omega}{(1+\tau_l^2\omega^2)(1+(1-F)\tau_l^2\omega^2)(\tau_l\omega)^{1/2}} F\frac{c_H}{c_l}\left(\frac{\tau_l}{\theta_l}\right)^{1/2}\right]; \tag{23}$$

$$\frac{\chi''}{\chi_0} = \left[\frac{F\tau_l\omega}{1+\tau_l^2\omega^2} - \frac{2F\tau_l\omega}{(1+\tau_l^2\omega^2)^2}\,\frac{c_H}{c_l}\right]\left[1 + \frac{1+(2(1-F)-F^2)\tau_l^2\omega^2+(1-F)^2\tau_l^4\omega^4}{F(1+\tau_l^2\omega^2)}\frac{\tau_s}{\tau_l} + \sqrt{0,3}\,\frac{1-\tau_l^2\omega^2+2\tau_l\omega}{(1+\tau_l^2\omega^2)(\tau_l\omega)^{3/2}}\,\frac{c_H}{c_l}\left(\frac{\tau_l}{\theta_l}\right)^{1/2}\right]. \tag{24}$$

The second and third terms, which are proportional to τ_s/τ_ℓ and $(c_H/c_\ell)(\tau_\ell/T_\ell)^{1/2}$ characterize the effect of spin relaxation and lattice relaxation on the susceptibility. If we estimate these effects for potassium chrome alum on the basis of the data in Refs. 3 and 14, at average fields ($F \sim 1$) these quantities become of the same order of magnitude at $T \sim 13°$ K, namely $10^{-4} - 10^{-5}$. Thus, when the effects of both relaxation processes on the complex susceptibility becomes equal they are no longer important.

9. In conlusion we shall use the results obtained in the present work for a qualitative discussion of a number of experiments carried out at helium temperatures[11,15,20,21] which have not been explained satisfactorily by the Casimir-Du Pré theory.[2] All these experiments were carried out in the frequency region for which $\tau_\ell\omega \sim 1$ and for average constant magnetic fields in the range ($10^3 - 10^4$ oersted). In this region of frequency and field strength relaxation effects are described by Eqs. (21) and (24). It is apparent from these equations that the role of lattice relaxation and spin relaxation are characterized respectively by the quantities $\theta_\ell c_H/\tau_\ell c_\ell$ and τ_s/τ_ℓ which we now evaluate.

As an example we consider iron ammonium alums. The quantity c_H is found from the formula[14] $c(b/c + H^2)T^{-2}$ where c and b are the constants in the Curie susceptibility relation and the specific heat of the spin system; $c = 8.8\times 10^{-3}$ deg-g^{-1} (Ref. 22), $b/c = 0.24 \times 10^6$ gauss^2 (Ref. 14). We take λ to be $1.31 \times 10^5\, T^3$ erg-sec^{-1}-deg^{-1} (Ref. 23) iron aommonium alum in the form of a pressed powder. The results given below for powder and crystal samples may differ from the true values but are correct within an order of magnitude.[18] Further, $\tau_\ell = (0.2 - 0.4) \times 10^{-3}$ sec for $H_0 = 285 - 968$ oersted and $T_0 = 2.13°$ K for the powder while $\tau_\ell = 7.5 \times 10^{-3}$ sec for $T_0 = 0.94°$ K in a field $H_0 = 748$ oersted for a single crystal;[11] $\rho = 1.7$ g-cm^{-3} (Ref. 22), and $\tau_s = 0.7 \times 10^{-9}$ sec (Ref. 14). Starting from these data, for a powder sample in the form of a sphere of radius 1 cm we obtain the following values for $\theta_\ell c_H/\tau_\ell c_\ell$: for $T_0 = 2.13°$ K and $H_0 = 285$ and 968 oersted, 0.32 and 0.53 respectively; for $T_0 = 0.93°$ K and $H_0 = 285$ and 968 oersted, 2.1 and 3.3 respectively. For a sample in the form of a single crystal an estimate gives the following values of $\theta_\ell c_H/\tau_\ell c_\ell$ for $H_0 = 748$ oersted: 0.02 for $T_0 = 2.16°$ K and 0.11 for $T_0 = 0.94°$ K. In the case of the single crystal with $T_0 = 2.16°$ K and $H_0 = 748$ oersted we have $\tau_s/\tau_\ell = 10^{-7}$. Thus the role of lattice relaxation is important in the materials being considered whereas spin relaxation is completely negligible.

A similar qualitative evaluation of experiments has been carried out for potassium chrome alums, cesium titanium alums, and manganese ammonium sulphate (of these materials the potassium chrome and cesium titanium alums have also been considered by Eisenstein[3,24]); the same qualitative results were obtained.

In conclusion we wish to express our gratitude to I. G. Shaposhnikov for proposing this work and for his guidance in the course of its execution.

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USE OF COLLECTIVE VARIABLES AND TREATMENT OF SHORT-RANGE FORCES IN THE THEORY OF A SYSTEM OF CHARGED PARTICLES

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The free energy and the distribution functions (binary and ternary) of a system of charged particles are calculated with effects of short-range forces included. Expressions for these quantities are written in terms of series of group integrals (correlations). It is shown that for an electron plasma in a compensating field the Coulomb potential does not give divergences in the expressions for the free energy and the distribution functions. The total free energy of a system of particles with a Coulomb interaction potential is also calculated. The "transition function" for such systems is constructed.

1. STATEMENT OF THE PROBLEM

UNTIL recently the determination of the thermodynamic characteristics of ionic systems has been carried out by the use of partial distribution functions — single-particle and binary functions. The calculations involved cumbersome computations and the solution of complicated systems of integro-differential equations. In these calculations for systems of charged particles short-range forces could be taken into account only with special forms of force law; for example, with the choice of the mutual potential in the form[1]

$$\Phi = (e^2/r)\,[1 - A\,(r)\,e^{-\varkappa r}].$$