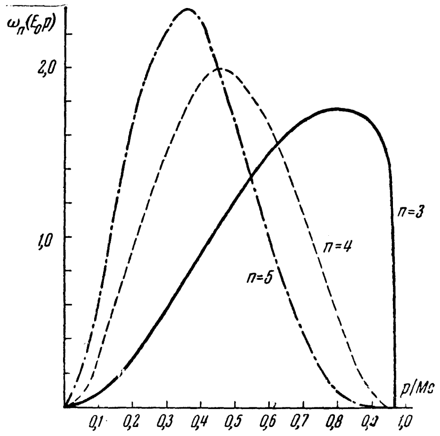

FIG. 1. Momentum distribution $\omega_n(E_0, p)$ of π-mesons for the process $N + N \rightarrow n\pi$ ($n = 3, 4, 5$). Here p is in units Mc = 0.93 Bev/c. The curves are normalized such that $\int \omega_n(E_0, p)\,dp = 1$.

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CONTRIBUTION TO THE PHENOMENOLOGICAL THEORY OF PARAMAGNETIC RELAXATION IN PARALLEL FIELDS

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The role of spin-lattice interaction in the phenomenological theory of complex paramagnetic susceptibility is taken into account in the case of parallel fields to a greater extent than was previously done in Ref. 3.

I. In the first work on the phenomenological theory of paramagnetic relaxation in parallel fields (Casimir and Du Pre[1] and others,(see Ref. 2) considered only the spin-lattice relaxation (see Ref. 2 for terminology). Later Shaposhnikov[3] (whose work will be designated hereafter by I) presented a phenomenological theory of complex paramagnetic susceptibility for the case of parallel fields, taking both spin-lattice and spin-spin relaxation into account, while Khutsishvili[4] has made a general phenomenological analysis of paramagnetic relaxation in a constant field using the Onsager principle, and has shown in particular under what assumptions the corresponding results of the theory given in I are obtained. Recently Yokota[5] repeated independently the examination of paramagnetic relaxation, previously carried out in I, generalizing somewhat the statement of the problem, and arriving at a final result (coinciding with the results of I) in only one particular case. In the present communication the theory of I is generalized with account of the work of Khutsishvili and Yokota. Here, as in I, we have in mind isotropic non-conducting paramagnetic materials in condensed state (for example, polycrystalline powders of paramagnetic salts, which are frequently used in experiments).

2. The theory of I is not general enough in the following two respects.

First, the expression given in I for the rate of change of the magnetic moment takes into account only the portion of this change that is effected by the interaction resulting in the spin-system from lack of internal equilibrium. In fact, however, the magnetic moment can change also by virtue of the action of the lattice on the spin system, provided only the various types of spin-lattice interactions include those involving the magnetic moment (for example, interaction by modulation of thermal oscillations of the spin-spin magnetic couplings or interaction through the spin-orbit couplings by modulation with thermal oscillations of the electric field of the lattice). This circumstance is not taken into account in the theory of I, making the latter suitable only for substances in which the change of the magnetic moment due to the spin-lattice interaction is sufficiently small compared with the change due to the internal interaction in the spin system. At room temperatures this is indeed so in many substances, as indicated by the experiments of Garif'ianov,[6] Sitnikov,[7] and Kurushin,[8] with which the theory of I is in good agreement, provided one takes into account the quantity τ_S, contained in I and called there the spin-spin relaxation time, which is independent of the value of the constant magnetic field. One must not however exclude the possibility of existence of substances in which one can no longer neglect the change in the magnetic moment due to the spin-lattice interaction. Calculations of the spin-lattice interaction made by Al'tshuler[9] for hydrated salts of rare-earth elements under the assumption that the basic mechanism of this interaction is the coupling between the spin, orbit, electric field, and thermal oscillations have shown that at room temperatures the spin-lattice relaxation time, defined as the reciprocal of the corresponding transition probability, is two orders shorter than the relaxation time of the magnetic moment due to the non-equilibrium state of the spin system and caused by the internal interaction in the spin system. With such a strong spin-lattice coupling one can expect that the portion of the change in the magnetic moment, due to the spin-lattice interaction, will also be substantial. Yokota[5] took this component of the change in magnetic moment into account, but it appears to us in a somewhat inconclusive manner.

Second, in the theory of I, as in all earlier phenomenological works on paramagnetic relaxation, the heat received by the spin system from the lattice was taken into account only by a term proportional to the difference in temperature between the spin system and the lattice. In the general case, however, the expression for the heat received by the spin system from the lattice should also contain a term due to the change in the magnetic moment under the influence of the spin-lattice interaction, as first pointed out by Khutsishvili.[4] In fact, the heat received by the spin system from the lattice is part of the change in the spin-system energy due to the action of the lattice on the spin system. But this action leads, generally speaking, to a change in both the spin-system temperature and the magnetic moment (see the remarks made above concerning spin-lattice interactions involving the magnetic moment), while the energy of the spin system depends in general on both these quantities. In particular cases the above-mentioned new term in the expression for the heat may be insignificant; this will occur if the spin-lattice interactions involving the magnetic moment are insignificant or if the energy of the spin system depends weakly on the magnetic moment.

The theory of I will be generalized below in the above two respects. This generalization consists, thus, of a more complete accounting for the role of the spin-lattice interaction.

3. As in I, we shall assume that the states (generally speaking not in equilibrium) of the spin system of a paramagnetic located in an external magnetic field of constant direction and magnitude

$$H = H_0 + \eta_0 e^{i\omega t}, \tag{1}$$

are fully determined by the temperature of the spin system T, by the magnetization M along the field (assume that the magnetization has no other components), and the intensity field H. In the presence of internal equilibrium in the spin system we would have

$$M = f(H, T), \tag{2}$$

but generally speaking there is no such relation. The lattice temperature will be assumed constant and equal to T_0.

Let us introduce the symbols

$$H - H_0 \equiv \eta, \quad T - T_0 \equiv \vartheta, \quad M - M_0 \equiv \xi, \tag{3}$$

where $M_0 = f(H_0, T_0)$ is the magnetization at full equilibrium of the paramagnetic in a constant field H_0 at a temperature T_0. The quantities η, ϑ, and ξ characterize the deviation of the paramagnetic from the state of complete equilibrium at H_0 and T_0; we shall assume the quantities small and linearize the equations of our problem relative to these quantities.

In the steady-state mode

$$\vartheta = \vartheta_0 e^{i\omega t}, \quad \xi = \xi_0 e^{i\omega t} \tag{4}$$

with complex amplitudes ϑ_0 and ξ_0. We have to find the complex magnetic susceptibility of the paramagnetic

$$\chi = \xi_0 / \eta_0. \tag{5}$$

For this purpose we use, as in I, the following two relations: an expression for the rate of change of the magnetization

$$\dot{M} = \dot{M}(H, T, M) \tag{6}$$

and the first law of thermodynamics for the spin system

$$dE = \delta Q + H dM, \tag{7}$$

where δ_Q is the heat received by the spin system from the lattice, and E is the energy of the spin system.

4. For the right half of (6) we use an expression given by Yokota

$$M = -\frac{1}{\tau_1}[M - f(H, T)] - \frac{1}{\tau_2}[M - f(H, T_0)], \tag{8}$$

where τ_1 and τ_2 are certain functions of H and T or of H and T_0 respectively. The form of these functions remains unknown within the framework of the phenomenological analysis. The first term characterizes here the change in the magnetic moment, occurring under the influence of the interaction in the spin system by virtue of the absence of internal equilibrium in the system, while the second term characterizes the change in the magnetic moment, occurring under the influence of the action of the lattice on the spin system. After linearization with respect to η, ϑ, and ξ Eq. (8) assumes the form

$$\dot{\xi} = -\frac{1}{\tau_1}\left(\xi - \frac{\partial f}{\partial T}\vartheta - \frac{\partial f}{\partial H}\eta\right) - \frac{1}{\tau_2}\left(\xi - \frac{\partial f}{\partial H}\eta\right), \tag{9}$$

where the quantities τ_1, τ_2 and the derivatives of f are taken at $H = H_0$ and $T = T_0$. The derivatives of f contained in (9) can be expressed in terms of the second derivatives of the thermodynamic potential $\Phi(H, T, N)$ of the spin system, taken at $H = H_0$, $T = T_0$, and $M = M_0$:

$$\partial f / \partial H = -\Phi_{MH} / \Phi_{MM}, \quad \partial f / \partial T = -\Phi_{MT} / \Phi_{MM}. \tag{10}$$

Furthermore, since $\partial f/\partial H$ is the isothermal equilibrium susceptibility χ_0, and $\Phi_{MH} = -1$ (because $\partial\Phi/\partial H = -M$), we get

$$\Phi_{MM} = \chi_0^{-1}, \tag{11}$$

and (10) can be rewritten

$$\partial f / \partial H = \chi_0, \quad \partial f / \partial T = -\chi_0 \Phi_{MT}. \tag{12}$$

Inserting (12) into (9) we obtain one of the equations needed for the determination of the quantity ξ_0, which is contained in formula (5) for the complex magnetic susceptibility

$$\tau_1\dot{\xi} + (\tau_1/\tau)\xi + \chi_0\Phi_{MT}\vartheta = \chi_0(\tau_1/\tau)\eta, \tag{13}$$

where

$$\tau = \tau_1\tau_2 / (\tau_1 + \tau_2). \tag{14}$$

Since it follows from (13) that it at $\eta = 0$ and $\vartheta = 0$

$$\tau\dot{\xi} + \xi = 0, \tag{15}$$

the quantity τ characterizes the rate at which equilibrium magnetization is established in a constant field at a constant temperature of the spin system, equal to the lattice temperature (for actual realization of such isothermal conditions for the spin system it is necessary that its coupling to the lattice be sufficiently strong). We shall call this quantity the time of isothermal relaxation of the magnetization (cf. Ref. 10). If the second term of the right half of Eq. (8) is assumed negligible, i.e., if $\tau_1/\tau_2 \ll 1$, we obtain by virtue of (14) in practice $\tau = \tau_1$, so that τ_1 is the time of isothermal relaxation of magnetization for that case, when the rate of change of the magnetic moment is determined primarily by the internal interaction in the spin system. Then (13) becomes, as it should, the corresponding equation of the theory of I, with τ_1 taking the role of the quantity τ_S, introduced in I and called there the spin-spin relaxation time. Thus, τ_1 coincides with the τ_S of I.

5. For δ_Q, which enters in (7), we assume the following expression

$$\delta Q = -\alpha\vartheta\, dt + \beta\,[M - f(H, T_0)]\, dt, \tag{16}$$

where α and β are functions of H_0 and T_0, which remain unknown in the phenomenological treatment, while dt is the time element; here the form of the second term, connected with the change of the magnetic moment under the influence of the spin-lattice interaction (see Sec. 2 above) is taken to conform with the form of the second term of the right half of (8).

Using the thermodynamic relations, it is possible to transform expression (7), after insertion of (16) and linearization with respect to η, ϑ, and ξ, and after making allowances for (11) and (12), to the following form

$$\rho_1\dot{\vartheta} + \gamma^{-1}\vartheta + \Phi_{TT}^{-1}\Phi_{MT}\,\rho_1\dot{\xi} - \chi_0^{-1}\gamma^{-1}\Phi_{MT}^{-1}[(\rho_1/\rho) - 1]\,\xi = -\gamma^{-1}\Phi_{MT}^{-1}[(\rho_1/\rho) - 1]\,\eta, \tag{17}$$

where

$$\gamma \equiv (1 - \chi_0\Phi_{TT}^{-1}\Phi_{MT}^2)^{-1}, \tag{18}$$

$$\rho = T_0\,(\chi_0\Phi_{MT}^2 - \Phi_{TT})\,(\alpha + \beta\chi_0\Phi_{MT})^{-1}, \tag{19}$$

$$\rho_1 = T_0\,(\chi_0\Phi_{MT}^2 - \Phi_{TT})\,\alpha^{-1}. \tag{20}$$

Equation (17) is the other equation needed to determine ξ_0.

Let the interaction in the spin system be so strong, that it causes internal equilibrium in the system to become established practically instantaneously ($\tau_1 = 0$), and let the external magnetic field remain constant ($\eta = 0$). We then obtain from (17) and (13), taking (14) into account,

$$\rho\dot{\vartheta} + \vartheta = 0, \tag{21}$$

so that the quantity ρ characterizes the rate at which the spin system and lattice temperatures become equalized in a constant field under that condition, that the spin system passes through a state of internal equilibrium. This quantity has been called spin-lattice relaxation time (see Refs. 1–3). If $\beta = 0$, i.e., if the energy exchange between the spin system and the lattice depends only on the difference in their temperatures, then, as can be seen from (19) and (20), the spin-lattice relaxation time is the quantity ρ_1, which coincides with the time of the spin-lattice relaxation employed in I, while Eq. (17) becomes in this case the corresponding equation in I, as should be.

6. To determine the complex susceptibility χ given by (5) it is necessary to insert (1) and (4) into Eqs. (13) and (17) and determine ξ_0 from these equations. We thus obtain

$$\frac{\chi}{\chi_0} = \frac{-1 + \rho_1/\rho + \tau_1/\tau + i\gamma\rho_1(\tau_1/\tau)\,\omega}{-1 + \rho_1/\rho + \tau_1/\tau - \gamma\rho_1\tau_1\omega^2 + i\,\{\rho_1 + \tau_1 + \gamma\rho_1[\tau_1/\tau - 1]\}\,\omega}\,. \tag{22}$$

If only the first terms are significant in (8) and (16), then $\tau_1/\tau \sim 1$ and $\rho_1/\rho \sim 1$, and (22) goes over into the formula for χ given in I with relaxation times ρ_1 and τ_1.

So far we have been unable to use the results of this general analysis to solve any particular problems in paramagnetic relaxation. It is possible, however, that these results will turn out to be useful in the evaluation of the recent experiments by Gorter and his associates[11] on paramagnetic absorption in parallel fields at high frequencies and at a temperature on the order of 20°K, in which maxima were observed in the curves for the absorption vs. the constant field at a given alternating field frequency, as contrasted with the monotonic decrease in this curve always observed in the previous experiments[6–8] at high and microwave frequencies at room temperatures.

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