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*ON THE THERMODYNAMICAL THEORY OF RELAXATION PHENOMENA IN SYSTEMS  
WITH ADDITIONAL PARAMETERS*

V. T. SHMATOV

Ural' Branch, Academy of Sciences, U.S.S.R.

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A thermodynamical theory of relaxation phenomena connected with the relaxation of an additional internal parameter of the system is developed on the basis of the method of Mandel'shtam and Leontovich. The relaxation times and the relation between them are established. Expressions are obtained for the dynamic derivatives of a system undergoing a periodic perturbation. A relation is found between the relaxation times and discontinuities in the derivatives near the Curie point. The results are extended to an arbitrary number of additional internal parameters.

THE term "additional internal parameter" is applied to a quantity characterizing some internal property of a system which, when the system is in equilibrium, appears to be a function of state. A delay on the part of the additional parameters in responding to external influences on the system leads, for example, to re-

laxation absorption of sound in liquids<sup>1,2</sup> or to internal friction in solids.<sup>3</sup> Below we develop further the method proposed by Mandel'shtam and Leontovich<sup>1</sup> for explaining acoustical absorption in liquids.

We consider a system characterized in its equilibrium state by a temperature  $T$ , a generalized force  $A$  (pressure, stress), and a coordinate  $a$  conjugate to it (volume, strain), connected by an equation of state; and some additional internal parameter  $\eta = \eta(T, a)$ . This parameter may, for example, be the degree of long- or short-range order, the spontaneous magnetization, the spontaneous electric polarization, the antiferromagnetic order, etc.

When the state of the system changes in time, the parameter  $\eta$  takes non-equilibrium values  $\eta \neq \eta(T, a)$ ; consequently, the system as a whole will pass through non-equilibrium states, in which it is described by the free energy  $F = F(T, a, \eta)$ . During this time we have  $F_{\eta}(T, a, \eta) \neq 0$ , where the subscript denotes, as usual, differentiation with respect to the corresponding parameter. When the system is in its equilibrium state,

$$F_{\eta}(T, a, \eta) = 0, \quad F_{\eta\eta} > 0. \quad (1)$$

According to Leontovich,<sup>4</sup> the entropy change of a non-equilibrium state is equal to

$$T dS = dU + A da - F_{\eta} d\eta,$$

where  $U(T, a, \eta)$  is the internal energy. Here the last term on the right determines the non-equilibrium part of the entropy change; we thus obtain for the rate of entropy production

$$T \Delta \dot{S} = -F_{\eta} \dot{\eta},$$

From this we have, in the approximation of the thermodynamics of irreversible processes,

$$\dot{\eta} = -L F_{\eta}(T, a, \eta). \quad (2)$$

Since  $\Delta \dot{S} > 0$ ,  $L > 0$ .

Expanding  $F_{\eta}$  in a series about the equilibrium state of the system and taking note of (1), we obtain from (2), in the linear approximation, an equation describing the time rate of change of the parameter  $\eta$ :

$$\tau^{aT} \dot{\eta} + \Delta \eta = \left( \frac{\partial \eta}{\partial a} \right)_T \Delta a + \left( \frac{\partial \eta}{\partial T} \right)_a \Delta T, \quad (3)$$

where  $\tau^{aT} = (L F_{\eta\eta})^{-1}$  is the relaxation time of the parameter  $\eta$  for constant  $a$  and  $T$ .

In order to go over to other variables in (3), we set  $A = -F_a$  and the entropy  $S = -F_T$ , so that to a linear approximation near the equilibrium state of the system

$$\Delta A = -F_{aa} \Delta a - F_{aT} \Delta T - F_{a\eta} \Delta \eta; \quad (4)$$

$$\Delta S = -F_{TT} \Delta T - F_{aT} \Delta a - F_{T\eta} \Delta \eta. \quad (5)$$

The derivatives  $F_{aa}$ ,  $F_{aT}$ , and  $F_{TT}$ , taken in the equilibrium state, can be expressed<sup>1</sup> in terms of the derivatives of the equilibrium state and  $F_{\eta\eta}$ :

$$F_{aa} = -\left( \frac{\partial A}{\partial a} \right)_T + \left( \frac{\partial \eta}{\partial a} \right)_T^2 F_{\eta\eta}; \quad F_{aT} = -\left( \frac{\partial A}{\partial T} \right)_a + \left( \frac{\partial \eta}{\partial a} \right)_T \left( \frac{\partial \eta}{\partial T} \right)_a F_{\eta\eta}; \quad T F_{TT} = -C_a + T \left( \frac{\partial \eta}{\partial T} \right)_a^2 F_{\eta\eta}, \quad (6)$$

where  $C_a$  is the specific heat of the system at constant  $a$ .

We have yet to explain the significance of the quantities entering into (6). If we exclude from our system the subsystem of the degrees of freedom responsible for the appearance of the properties characterized by the parameter  $\eta$ , i.e., if we set in (6)  $(\partial \eta / \partial a)_T \equiv (\partial \eta / \partial T)_a \equiv 0$ , then it becomes clear that, in the system remaining after the exclusion of the subsystem,  $-F_{aa}$ ,  $-F_{aT}$ , and  $-T F_{TT}$  represent the derivatives  $(\partial A / \partial a)_T$ ,  $(\partial A / \partial T)_a$ , and the specific heat  $C'_a$ . Consequently, in turn, the quantities

$$\left[ \left( \frac{\partial A}{\partial a} \right)_T \right] = \left( \frac{\partial \eta}{\partial a} \right)_T^2 F_{\eta\eta}; \quad \left[ \left( \frac{\partial A}{\partial T} \right)_a \right] = \left( \frac{\partial \eta}{\partial a} \right)_T \left( \frac{\partial \eta}{\partial T} \right)_a F_{\eta\eta}; \quad [C_a] = T \left( \frac{\partial \eta}{\partial T} \right)_a^2 F_{\eta\eta} \quad (7)$$

are the respective contributions of the subsystem to  $(\partial A / \partial a)_T$ ,  $(\partial A / \partial T)_a$ , and to the specific heat  $C_a$  of the whole system. In particular, if a second-order phase transition connected with the existence of the parameter  $\eta$ , is possible for our system, then at the Curie point  $[(\partial A / \partial a)_T]$ ,  $[(\partial A / \partial T)_a]$ , and  $[C_a]$  will be equal to the discontinuities of the corresponding quantities.

Using Eqs. (4) and (5) to transform (3) successively to pairs of variables other than  $a$  and  $T$ , we

can show that the time rate of change of  $\eta$ , for an arbitrary choice of variables, has the form

$$\tau^{xy} \dot{\eta} + \Delta\eta = (\partial\eta/\partial x)_y \Delta x + (\partial\eta/\partial y)_x \Delta y, \quad (8)$$

where  $x$  and  $y$  are any two of the variables  $a$ ,  $A$ ,  $T$ , and  $S$ . Here the relaxation times  $\tau^{xy}$  of the parameter  $\eta$ , for constant  $x$  and  $y$ , are related to each other by

$$\begin{aligned} \tau^{aT} &= \tau^{AT} \frac{(\partial A/\partial a)_T}{(\partial A/\partial a)_T - [(\partial A/\partial a)_T]} = \tau^{Aa} \frac{(\partial A/\partial T)_a}{(\partial A/\partial T)_a - [(\partial A/\partial T)_a]} = \tau^{TS} \frac{(\partial A/\partial T)_a}{(\partial A/\partial T)_a - [(\partial A/\partial T)_a]} = \tau^{aS} \frac{C_a}{C_a - [C_a]} \\ &= \tau^{AS} \frac{(\partial A/\partial a)_T C_A}{\{(\partial A/\partial a)_T - [(\partial A/\partial a)_T]\} (C_A - [C_A])}. \end{aligned} \quad (9)$$

In deriving (9), use has been made of the equations in (6) and the notation of (7).

For a periodic perturbation of the system at a frequency  $\omega$ , we can find the dynamic derivatives  $(\partial y/\partial x)_{a,\omega}$ , where  $x$ ,  $y$ ,  $z$  are any three of the variables  $a$ ,  $A$ ,  $T$ , or  $S$ . Eliminating  $\Delta\eta$  from two equations of type (8), expressed in terms of the variables  $x$ ,  $z$  and  $y$ ,  $z$ , we obtain

$$\left(\frac{\partial y}{\partial x}\right)_{z,\omega} = \left(\frac{\partial y}{\partial x}\right)_z \frac{1 + i\omega\tau^{yz}}{1 + i\omega\tau^{xz}}. \quad (10)$$

From this it follows that

$$\left(\frac{\partial y}{\partial x}\right)_{z,\omega=\infty} : \left(\frac{\partial y}{\partial x}\right)_z = \tau^{yz} : \tau^{xz}. \quad (11)$$

For the variables  $P$  (pressure),  $V$  (volume),  $T$ , and  $S$ , the equations obtained by Davis and Lamb by another method<sup>6</sup> follow from (11) immediately.

From (8) it follows that

$$(\partial\eta/\partial x)_{y,\omega=\infty} \equiv (\partial\eta/\partial x)_{y,\omega=\infty} \equiv 0,$$

so that, for  $\omega \rightarrow \infty$ , the subsystem of the degrees of freedom responsible for the appearance of the system properties characterized by the parameter  $\eta$  take no part in the change of the state of the system; it is isolated from the remaining degrees of freedom of the whole system. Therefore the quantities  $(\partial y/\partial x)_{z,\omega=\infty}$  are just the derivatives  $(\partial y/\partial x)'_z$  relative to the system which remains after the subsystem has been excluded, and the quantity  $[(\partial y/\partial x)_z] = (\partial y/\partial x)_z - (\partial y/\partial x)'_z$  determines the contribution of the subsystem to the derivative  $(\partial y/\partial x)_z$  for the total system. On this basis we obtain from (11)

$$\tau^{yz}/\tau^{xz} = 1 - [(\partial y/\partial x)_z] / \left(\frac{\partial y}{\partial x}\right)_z. \quad (12)$$

At the Curie point,  $[(\partial y/\partial x)_z] = \Delta(\partial y/\partial x)_z$ ; consequently, one can write, near enough to the Curie point

$$\frac{\tau^{yz}}{\tau^{xz}} = 1 - \Delta \left(\frac{\partial y}{\partial x}\right)_z \bigg/ \left(\frac{\partial y}{\partial x}\right)_z. \quad (13)$$

At the Curie point the relaxation times increase without limit.<sup>7</sup>

The results obtained above can be extended to a system with an arbitrary number  $N$  of additional internal parameters  $\eta_1, \dots, \eta_N$  by an analogous scheme. In this case we have for the dynamic derivatives

$$\left(\frac{\partial y}{\partial x}\right)_{z,\omega} = \left(\frac{\partial y}{\partial x}\right)_z \prod_{n=1}^N \frac{1 + i\omega\tau_n^{yz}}{1 + i\omega\tau_n^{xz}}, \quad (14)$$

where  $\tau_n^{yz}$  and  $\tau_n^{xz}$  are the relaxation times of the additional parameters for constant  $y$ ,  $z$  and constant  $x$ ,  $y$  respectively. These times relate to the additional parameters with the same index  $n$ , provided that  $\eta_1, \dots, \eta_N$  are independent. In the opposite case the relaxation times appear to be general ones and cannot be associated with individual parameters. In particular, the approach of the  $\eta_n$  to their equilibrium values  $\eta_n^{(0)}$  for constant  $x$ ,  $y$  will take place according to the law

$$\eta_n = \eta_n^{(0)} + \sum_{k=1}^N A_{nk}^{xy} \exp(-t/\tau_k^{xy}), \quad (15)$$

where the coefficients  $A$  depend on the initial conditions.

Equations (10) and (11) can be applied to the description of all relaxation phenomena connected with the corresponding dynamic derivatives. The thermodynamical theory leaves open the question of the temperature dependence of the relaxation times.

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## A POSSIBLE TEST OF THE CONSERVATION OF "COMBINED PARITY" IN THE BETA INTERACTION

A. Z. DOLGINOV

Leningrad Physico-Technical Institute, Academy of Sciences, U.S.S.R.

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It is suggested that the invariance of the  $\beta$ -interaction under time reversal can be examined by observing  $\beta-\gamma$  angular correlations in allowed transitions with aligned nuclei. Formulas are obtained for the  $\beta-\gamma$  correlation in nuclei oriented by various methods. The general form of the  $\beta$ -interaction is considered with parity nonconservation taken into account. An expression is given for the  $\beta-\gamma$  angular and polarization correlations in oriented nuclei for  $\beta$ -transitions of any order of forbiddenness.

**EXPERIMENT** has at present verified that parity is not conserved in weak interactions, as had been hypothesized in different forms by Lee and Yang<sup>1,2</sup> and by Landau.<sup>3,4</sup> It remains for experiment to test the specific versions of the hypothesis. We shall speak here primarily of a test of the conservation law for "combined parity," which according to Pauli's theorem<sup>5</sup> is equivalent to the invariance under time reversal.

Another important question is whether or not it is possible to describe the neutrino by a two-component equation.<sup>4,2</sup>

In the present work we shall give our main consideration to the first of these problems. We shall do this by investigating the  $\beta-\gamma$  angular correlation of oriented nuclei. We shall show that if the nuclei are oriented by the method of Bleaney<sup>6</sup> or Pound<sup>7</sup> (aligned nuclei), then a measurement of the  $\beta-\gamma$  correlation for allowed transitions can give important information on the nature of the  $\beta$ -interaction:

(1) If there is no  $\beta-\gamma$  angular correlation, then (a) the  $\beta$ -interaction is invariant with respect to