

# Establishment of spin temperature and the theory of magnetic resonance saturation

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(Submitted April 10, 1975; resubmitted September 25, 1975)

Zh. Eksp. Teor. Fiz. 70, 560-568 (February 1976)

The establishment of the spin temperature is considered by using the nonequilibrium statistical operator technique. It is shown that the dependence of the rate at which a single temperature of the secular part of the spin-spin interactions and a unified pool of the Zeeman energy and the energy of the nonsecular spin-spin interactions are established on the resonance frequency  $\omega_0$  is Lorentzian providing  $\omega_0$  exceeds several resonance widths. The theoretical and experimental results on the establishment of a single temperature in the spin system are analyzed. The magnetic resonance equations are written down by taking into account the relation between the dipole-dipole pool and Zeeman pool due to the nonsecular part of the spin-spin interactions. An expression for the power absorbed from the alternating field is obtained. The feasibility of experimental investigations of the role of nonsecular interactions in magnetic resonance absorption is discussed.

PACS numbers: 33.90.+h

## 1. INTRODUCTION

In investigations of the role of the singular part of spin-spin interactions (dipole-dipole pool-DDP) in magnetic resonance it is customary to neglect the coupling between the DDP and the Zeeman pool (the  $z$ -pool) due to the nonsecular dipole-dipole interactions (NDDI).<sup>[1,2]</sup> This neglect is based on the assumption that a change  $\Delta E_z$  in the energy of the  $z$ -pool is possible in the absence of interaction with the external alternating field  $H_r$  and the lattice only if an equal change  $\Delta E_d$  takes place in the DDP energy. Mathematically this is expressed, in particular, in the fact that the operators in the corresponding correlation functions are written in the interaction representation in the NDDI.

It follows from this approach that the connection between the DDP and the  $z$ -pool is effective only if the  $H_0$  does not exceed several times the local field  $H_{loc}$ . In this case one ignores the fact that the NDDI is an energy tank through which energy is exchanged between the DDP and the  $z$ -pool. Since the NDDI is an energy tank, energy transitions in the spin system become possible, through which change of the energy of the DDP is not equal to the change in the energy of the  $z$ -pool. The energy conservation law is satisfied in this case because of the change in the energy of the NDDI. In the course of such an energy exchange, the temperature of the DDP and of the unified pool of Zeeman energy and the NDDI energy, which we shall henceforth call subsystem 1, changes (equalization of the temperatures of the  $z$ -pool and of the NDDI within a time on the order of  $T_2$  is a fact convincingly demonstrated theoretically and experimentally).<sup>[2]</sup>

In this paper, using the method of nonequilibrium statistical operator (NSO),<sup>[3]</sup> we consider the establishment of a unified temperature of the subsystem 1 and of the DDP, and investigate the role of the NDDI in the theory of magnetic resonant absorption.

## 2. EQUALIZATION OF THE TEMPERATURE OF THE SUBSYSTEM 1 AND OF THE DDP

We separate in the Hamiltonian of the investigated system the secular and the nonsecular terms:

$$\mathcal{H} = \mathcal{H}_z + \mathcal{H}_{sec} + \mathcal{H}_{n, sec}, \quad (1)$$

where

$$\begin{aligned} \mathcal{H}_z &= \hbar \omega_0 \sum_i S_i^z, \\ \mathcal{H}_{sec} &= \sum_{i,j} [A_{ij} S_i^+ S_j^+ + B_{ij} (S_i^+ S_j^- + S_i^- S_j^+)], \\ \mathcal{H}_{n, sec} &= C + C^+ + \mathcal{E} + \mathcal{E}^+, \\ C &= \sum_{i,j} C_{ij} S_i^+ S_j^+, \quad \mathcal{E} = \sum_{k,l} E_{kl} S_k^+ S_l^+. \end{aligned}$$

Goldman<sup>[2]</sup> used the operator

$$R = \frac{i(C^+ - C)}{\omega_0} + \frac{i(\mathcal{E}^+ - \mathcal{E})}{2\omega_0} \quad (2)$$

to effect the transition from the Hamiltonian (1) to the Hamiltonian

$$\mathcal{H}' = Z^* + \mathcal{H}_{sec}^* + V, \quad (3)$$

where

$$\begin{aligned} Z^* &= \mathcal{H}_z + \mathcal{H}_{n, sec}, \quad \mathcal{H}_{sec}^* = \mathcal{H}_{sec}^* - i[R, \mathcal{H}_{sec}], \\ V &= i[R, \mathcal{H}_{sec}^*]. \end{aligned}$$

It is stated there that the operators  $Z^*$  and  $\mathcal{H}_{sec}^*$  commute with each other, and the operator  $V$  commutes neither with  $Z^*$  nor with  $\mathcal{H}_{sec}^*$  and is a perturbation that ensures the coupling of the energy pools corresponding to  $Z^*$  and  $\mathcal{H}_{sec}^*$ .

Further, Goldman<sup>[2]</sup> wrote down, for the case of high temperatures, on the basis of expansion of the density matrix up to second order in the perturbation  $V$ , equations for the reciprocal temperatures  $\alpha$  and  $\beta$  of the subsystems  $Z^*$  and  $\mathcal{H}_{sec}^*$ . Accordingly, the probabilities are calculated by Goldman in the representation of the interaction in  $V$ , which is equivalent to the representation of the interaction in the NDDI (cf. formulas (6.31) and (6.37) in<sup>[2]</sup>). As the result, he took into ac-

count in<sup>[2]</sup> only those transitions in the spin system in which the change of the DDP energy is completely compensated by the change of the Zeeman energy. The interconnection of the NDDI with the  $z$ -pool affects in this case only the heat capacity of the subsystem 1.

The equations for the reciprocal temperatures  $\alpha$  and  $\beta$  in<sup>[2]</sup> are based on the statement that the commutator  $[Z^*, \mathcal{H}_{\text{sec}}^*]$  is equal to zero. By direct calculations it can be verified that  $[Z^*, \mathcal{H}_{\text{sec}}^*] = -i[\mathcal{H}_{\text{nsec}}, [R, \mathcal{H}_{\text{sec}}]]$ .

Thus, the commutator  $[Z^*, \mathcal{H}_{\text{sec}}^*]$  is indeed small in comparison with  $Z^*$  and  $\mathcal{H}_{\text{sec}}^*$ . On this basis, the operators  $Z^*$  and  $\mathcal{H}_{\text{sec}}^*$  in<sup>[2]</sup> are regarded as commuting. However, the commutator  $[\mathcal{H}_{\text{sec}}^*, V]$ , which causes the coupling of the reservoirs  $Z^*$  and  $\mathcal{H}_{\text{sec}}^*$  via the interaction  $V$ , is a quantity of the same order as  $[Z^*, \mathcal{H}_{\text{sec}}^*]$ . Consequently the assumption that the coupling between the pools  $Z^*$  and  $\mathcal{H}_{\text{sec}}^*$  is due only to the perturbation  $V$  can lead to erroneous results. No proof whatever is presented in<sup>[2]</sup> that the quantity  $[Z^*, \mathcal{H}_{\text{sec}}^*]$  can be neglected against the background of  $[\mathcal{H}_{\text{sec}}^*, V]$ . The separation in the DDP of a principal part commuting with the NDDI remains physically unexplained in our opinion.

The NSO method<sup>[3]</sup> makes it possible to write down equations for the reciprocal temperatures  $\beta_1$  and  $\beta_2$  of the subsystem 1 and of the DDP, without resorting to any separation in the DDP or to expansion of the density matrix in terms of the interaction. The probabilities are calculated in this case in the Heisenberg representation, making it possible to take into account in the spin system transitions in which the cancellation of the change in the DDP energy is ensured on account of the NDDI energy.

As already indicated in the Introduction, after a time on the order of  $T_2$  the subsystem 1 is produced, and we then deal with a system consisting of subsystem 1 and the DDP. We write down the NSO of such a system, using the form<sup>[3]</sup>, p. 401

$$\rho = \varepsilon \int_{-\infty}^0 e^{t'} \rho_q(t+t', t') dt', \quad (4)$$

$$\rho_q(t+t', t') = \exp\left(-\frac{i}{\hbar} \mathcal{H} t'\right) \rho_q(t+t') \exp\left(-\frac{i}{\hbar} \mathcal{H} t'\right), \quad (5)$$

with the quasilocal operator in the form

$$\rho_q(t+t') = \exp\left\{-\Phi(t+t') - \sum_m P_m F_m(t+t')\right\}, \quad (6)$$

$$\Phi(t+t') = \ln \text{Sp} \exp\left\{-\sum_m P_m F_m(t+t')\right\}.$$

For high temperatures we have

$$\rho_q(t+t') = \frac{1}{\text{Sp} 1} \left[1 - \sum_m P_m F_m(t+t')\right] \quad (7)$$

and

$$\rho = \frac{1}{\text{Sp} 1} \left\{1 - \sum_m P_m F_m(t) + \int_{-\infty}^0 dt' e^{t'} \frac{d}{dt'} \sum_m F_m(t+t') P_m(t')\right\}. \quad (8)$$

In our case

$$P_1 = \mathcal{H}_1 = \mathcal{H}_z + \mathcal{H}_{\text{nsec}}, \quad P_2 = \mathcal{H}_2 = \mathcal{H}_{\text{sec}},$$

and  $F_1 = \beta_1$  and  $F_2 = \beta_2$ .

Putting

$$\langle \dots \rangle = \text{Sp}(\dots) / \text{Sp} 1, \quad \bar{Q} = \text{Sp}(\rho Q)$$

and bearing in mind that

$$\frac{d\beta_i}{dt} \langle \mathcal{H}_i^2 \rangle = \frac{i}{\hbar} [\mathcal{H}_i, \mathcal{H}], \quad (9)$$

we obtain with the aid of (8)

$$\frac{d\beta_1}{dt} \langle \mathcal{H}_1^2 \rangle = -\frac{i}{\hbar} \sum_m \int_{-\infty}^0 e^{t'} \frac{d}{dt'} \beta_m(t+t') \langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] \mathcal{H}_m(t') \rangle dt', \quad (10)$$

$$\frac{d\beta_2}{dt} \langle \mathcal{H}_2^2 \rangle = \frac{i}{\hbar} \sum_m \int_{-\infty}^0 e^{t'} \frac{d}{dt'} \beta_m(t+t') \langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] \mathcal{H}_m(t') \rangle dt.$$

Bearing in mind the fact that  $\beta_1(t)$  is a slow function of the time in comparison with  $\langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] \mathcal{H}_m(t) \rangle$ , we take  $\beta(t)$  outside the signs of differentiation and integration with respect to  $t'$ . Equation (10) then takes the form

$$d\beta_1/dt = (\beta_2 - \beta_1) W_1, \quad d\beta_2/dt = (\beta_1 - \beta_2) W_2, \quad (11)$$

where

$$W_1 = L_{21} / \langle \mathcal{H}_1^2 \rangle, \quad W_2 = L_{21} / \langle \mathcal{H}_2^2 \rangle; \quad (12)$$

$$L_{21} = \frac{-1}{\hbar^2} \int_{-\infty}^0 e^{t'} \langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t) \rangle dt = \frac{-1}{2\hbar^2} J(0), \quad (13)$$

$$J(\omega) = \int_{-\infty}^{\infty} \langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t) \rangle e^{-i\omega t} dt. \quad (14)$$

Let us estimate the value of  $J(0)$  for fields  $H_0$  exceeding several values of  $H_{10c}$ . We put

$$\langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t) \rangle = G(t). \quad (15)$$

As is well known

$$G(t) = G(t_1 - t_2) = \langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t_1) [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t_2) \rangle, \quad (16)$$

if  $t_1 - t_2 = t$ .

Writing on the equations for the Fourier transform

$$\langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t_1) [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t_2) \rangle_0$$

of the correlation function (16) using differentiation with respect to both arguments  $t_1$  and  $t_2$ , we obtain, after neglecting inessential terms for the values  $H_0 \gg H_{10c}$ :

$$J(0) = \langle [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}] [\mathcal{H}_2, \mathcal{H}_{\text{nsec}}](t) \rangle_0 = \frac{1}{\hbar^2 \omega_0^2} [A_1(0) + i A_2(0)], \quad (17)$$

where

$$A_1(\omega) = \langle [\mathcal{H}_2, [C^+, C]](t_1) [\mathcal{H}_2, [C^+, C]](t_2) \rangle_0, \quad (18)$$

$$A_2(\omega) = \langle [\mathcal{H}_2, [\mathcal{E}^+, \mathcal{E}]](t_1) [\mathcal{H}_2, [\mathcal{E}^+, \mathcal{E}]](t_2) \rangle_{\omega} \quad (19)$$

It is seen from (17) that the  $J(\omega)$  curve has a satellite in the vicinity of the frequency  $\omega = 0$ . This satellite is the consequence of the presence of nonsecular terms in the Hamiltonian and reflects the possibility of energy transitions in the spin system, such that the change in the energy of the DDP is offset by the change of the energy of NDDI.

Thus, to determine  $J(0)$  it is necessary to find  $A_1(0)$  and  $A_2(0)$ . This could be done by finding the forms of the curves  $A_1(\omega)$  and  $A_2(\omega)$ . But the determination of the forms of these curves on the basis of the existing methods calls for cumbersome calculations and is a problem much more complicated than the calculation of the form of the absorption curve or the  $\langle S^z S^z(t) \rangle_{\omega}$  curve. At the same time, if the  $g$  factor is weakly anisotropic, then simple estimates with which to determine the order of magnitude of  $A_1(0)$  and  $A_2(0)$  seem possible to us. Indeed, the widths of the lines  $A_1(\omega)$  and  $A_2(\omega)$  is close to the width of the magnetic resonance line  $\Delta\omega = 2\pi T_2^{-1}$ . Next, for  $A_1(\omega)$  we have

$$\int_{-\infty}^{\infty} A_1(\omega) d\omega = 2\pi \langle [\mathcal{H}_2, [C^+, C]] [\mathcal{H}_2, [C^+, C]] \rangle = 2\pi A_{10} \quad (20)$$

We put

$$A_1(0) = \frac{1}{\Delta\omega} \int_{-\infty}^{\infty} A_1(\omega) d\omega = \frac{A_{10}}{T_2^{-1}} \quad (21)$$

Assuming  $A_1(\omega)$  to have a Gaussian or Lorentzian shape while retaining the normalization (20), we can easily verify with the aid of simple calculations that the  $A_1(0)$  is in either case very close to  $A_{10}/T_2^{-1}$ . Analogously, for estimates of  $A_2(0)$  we can use the formula

$$A_2(0) = A_{20}/T_2^{-1}, \quad (22)$$

where

$$A_{20} = \langle [\mathcal{H}_2, [\mathcal{E}^+, \mathcal{E}]] [\mathcal{H}_2, [\mathcal{E}^+, \mathcal{E}]] \rangle.$$

The values of  $A_{10}$  and  $A_{20}$  can be calculated directly, but the resultant cumbersome expressions are not convenient for use. At the same time it is obvious that to estimate the probability  $W_2$  we can use the expression

$$W_2 = -J(0)/2\hbar^2 \langle \mathcal{H}_2^2 \rangle \approx 1/\tau^3 \omega_0^2, \quad (23)$$

where  $\tau$  is of the order of  $T_2$ .

If the  $g$  factor is strongly anisotropic, then formula (23) for the determination of  $W_2$  is not valid. In this case, however, reasonable estimates based on allowance for the dependence of  $A_{10}$ ,  $A_{20}$ , and  $\langle \mathcal{H}_2^2 \rangle$  on  $g_{\parallel}$  and  $g_{\perp}$  are possible. We shall henceforth assume the  $g$  factor to be weakly anisotropic and use formula (23) for  $W_2$ .

The solution of (11) takes the form

$$\beta_1 = C_1 e^{-t/\tau} + \beta_e, \quad \beta_2 = -C_1 \frac{\langle \mathcal{H}_1^2 \rangle}{\langle \mathcal{H}_2^2 \rangle} e^{-t/\tau} + \beta_e, \quad (24)$$

where

$$T^{-1} = W_1 + W_2, \quad W_1 = W_2 \langle \mathcal{H}_2^2 \rangle / \langle \mathcal{H}_1^2 \rangle. \quad (25)$$

It is seen from (23) that the subsystem 1 and the DDP turn out to be sufficiently strongly coupled as a result of the NDDI in the case when  $H_0 \gg H_{10c}$  (of course, not strongly enough to make the concept of two spin temperatures inapplicable).

The Lorentzian character of the dependence of  $T^{-1}$  on  $\omega_0$  manifests itself for regular magnets in fields  $H_0$  exceeding certain values  $H_{10c}$ . This explains the apparent contradiction between the result of the present paper and the experimental investigations of the dependence of  $T^{-1}$  on  $H_0$ . In<sup>[2]</sup>, on p. 212 (of the Russian translation) is given the result of the measurements made by Hartmann and Anderson<sup>[4]</sup> of the dependence of  $T^{-1}$  on  $H_0$  at the  ${}^7\text{Li}$  nuclei in metallic lithium. The Gaussian dependence of  $T^{-1}$  on  $H_0$  was observed at values of  $H_0$  from 10 to 15 Oe, that is, within the limits of two or three resonance widths. In fields exceeding 15 Oe, a noticeable decrease of the rate of the dependence of  $T^{-1}$  on  $H_0$  was observed. The value of  $T$  at  $H_0 > 15$  Oe, calculated with the aid of formula (23) at values  $\tau \approx 1.2 \times 10^{-3}$  sec close to the value  $T_2 \approx 0.5 \times 10^{-3}$  sec for  ${}^7\text{Li}$ , agree well with experiment.

In Caspers' book,<sup>[5]</sup> the Gaussian dependence of the probability of the longitudinal spin-spin relaxation of  $T^{-1}$  on  $H_0$  is postulated by starting from the shape of the unsaturated resonance curve in concentrated magnets. The data of Locher and Verstelle<sup>[6]</sup> for Tutton's salt, pertaining to fields  $H_0$  in the interval from 1100 to 2000 Oe at a resonance-curve width 1460 Oe are cited in<sup>[5]</sup> as a confirmation of the Gaussian dependence of  $T^{-1}$  on  $H_0$ . At these values of  $H_0$ , the satellite of the  $J(\omega)$  cannot determine the value of  $T^{-1}$ .

In spite of a careful search of the literature, I was unable to find experimental proof of a Gaussian dependence of the probability of  $T^{-1}$  on  $H_0$  at  $H_0 \gg H_{10c}$ . The absence of measurement data on  $T$  in strong field ( $H_0 \gg H_{10c}$ ) is apparently due both to difficulties of experimental nature and to the absence of a satisfactory theoretical interpretation of the measurements in the case when the dependence of  $T$  on  $H_0$  deviates from Gaussian.

### 3. MAGNETIC RESONANCE SATURATION

We examine now the role of the NDDI in the absorption by the spin system of the energy of the external alternating field.

The Provotorov equations,<sup>[1]</sup> which take into account the role of the DDP in the theory of resonant absorption, can be obtained by the NSO method<sup>[3]</sup> if, following Buishvili,<sup>[7]</sup> the external field is regarded as a subsystem with a zero reciprocal temperature. This approach, which includes the dependence of the Hamiltonian on the time, was used by Al'tshuler *et al.*,<sup>[8]</sup> with Eqs. (10) of<sup>[8]</sup> coinciding with the Provotorov equations<sup>[1]</sup> in the particular case  $z = 0$ . In both<sup>[8]</sup> and<sup>[1]</sup>, however, the coupling of the DDP with the  $z$ -pool via the NDDI is neglected.

We derive equations for the absorption of the energy of an external alternating field  $H_r$  with allowance for the role of the NDDI. As the quasi-independent subsystems we consider the subsystem 1 and the DDP or the subsystem 2 and subsystem 3 with Hamiltonian

$$\mathcal{H}_s = \mathcal{H}_r + \mathcal{H}_{sr}, \quad (26)$$

where the Hamiltonian of the external field is

$$\mathcal{H}_r = \hbar\omega_r B^+ B \quad (27)$$

and the Hamiltonian of the spin-photon interaction is

$$\mathcal{H}_{sr} = V_1 \sum_i (S_i^+ + S_i^-) (B^+ + B). \quad (28)$$

The total Hamiltonian of the system is

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3. \quad (29)$$

The equations for  $\beta_1$  and  $\beta_2$  in the presence of an external alternating field are obtained with the aid of an NSO in the form (8), in analogy with Eqs. (11), taking the spin-lattice relaxation (SLR) into account phenomenologically:

$$\begin{aligned} \langle \mathcal{H}_1^2 \rangle \frac{d\beta_1}{dt} &= -\beta_1 (L_{21} + L_{31} + L_{sL}^{(1)}) + \beta_2 (L_{21} - \alpha_1 L_{31}) + \beta_0 L_{sL}^{(1)}, \\ \langle \mathcal{H}_2^2 \rangle \frac{d\beta_2}{dt} &= -\beta_2 (L_{21} + \alpha_1^2 L_{31} + L_{sL}^{(2)}) + \beta_1 (L_{21} - \alpha_1 L_{31}) + \beta_0 L_{sL}^{(2)}. \end{aligned} \quad (30)$$

Here

$$\alpha_1 = (\omega_1 - \omega_0) / \omega_0, \quad L_{31} = \langle \mathcal{H}_2^2 \rangle V_1^2 \langle B^+ B + 1 \rangle f(\omega_1 - \omega_0), \quad (31)$$

where  $f(\omega - \omega_0)$  is a function of the shape of the unsaturated absorption curve,

$$L_{sL}^{(1)} = T_{1(1)}^{-1} \langle \mathcal{H}_1^2 \rangle, \quad L_{sL}^{(2)} = T_{1(2)}^{-1} \langle \mathcal{H}_2^2 \rangle, \quad (32)$$

with  $T_{1(1)}$  and  $T_{1(2)}$  respectively the SLR times of substances 1 and 2.

We rewrite (30) in the form

$$d\beta_1/dt = a_1\beta_1 + b_1\beta_2 + c_1, \quad d\beta_2/dt = a_2\beta_1 + b_2\beta_2 + c_2. \quad (33)$$

The solution of Eqs. (33) takes the form

$$\begin{aligned} \beta_1 &= A_1 e^{s_1 t} + A_2 e^{s_2 t} + \beta_{1c}, \\ \beta_2 &= A_1 \frac{s_1 - a_1}{b_1} e^{s_1 t} + A_2 \frac{s_2 - a_1}{b_1} e^{s_2 t} + \beta_{2c}, \end{aligned} \quad (34)$$

where

$$s_{1,2} = \frac{1}{2} \{ a_1 + b_2 \pm [(a_1 - b_1)^2 + 4b_1 a_2]^{1/2} \}, \quad (35)$$

$$\beta_{1c} = \frac{b_1 c_2 - b_2 c_1}{b_2 a_1 - b_1 a_2}, \quad \beta_{2c} = \frac{a_2 c_1 - a_1 c_2}{b_2 a_1 - b_1 a_2}. \quad (36)$$

It is seen from (34) that in the stationary case  $\beta_1 = \beta_{1c}$  and  $\beta_2 = \beta_{2c}$ .

With the aid of (30) we find that the change of the en-

ergy of the spin system due to the alternating field or, in other words, the absorbed power of the alternating field, is

$$P_r = L_{31} (1 + \alpha_1) (\beta_1 + \alpha_1 \beta_2). \quad (37)$$

In the stationary case, substituting in (36) the expressions for the coefficients  $a$ ,  $b$ , and  $c$  in terms of  $L$ , we obtain

$$P_{rc} = \frac{\beta_0 L_{31} (1 + \alpha_1)^2 [L_{21} (L_{sL}^{(1)} + L_{sL}^{(2)}) + L_{sL}^{(1)} L_{sL}^{(2)}]}{L_{21} [L_{31} (1 + \alpha_1)^2 + L_{sL}^{(1)} + L_{sL}^{(2)}] + L_{31} [L_{sL}^{(2)} + \alpha_1^2 L_{sL}^{(1)}] + L_{sL}^{(1)} L_{sL}^{(2)}}. \quad (38)$$

In the case  $L_{21} \ll L_{sL}^{(2)}$  we get from (38)

$$P_{rc} = \beta_0 L_{31} (1 + \alpha_1)^2 / \left[ 1 + \frac{L_{31}}{L_{sL}^{(1)}} \left( 1 + \alpha_1^2 \frac{L_{sL}^{(1)}}{L_{sL}^{(2)}} \right) \right]. \quad (39)$$

Formula (39) differs from the result of Provotorov<sup>[1]</sup> by the factor  $(1 + \alpha_1)^2$ , which takes into account the energy absorption due to the DDP. In weak fields at large detunings  $\omega_1 - \omega_0$ , the factor  $(1 + \alpha_1)^2$  must be taken into consideration.

At  $L_{21} \gg L_{sL}^{(2)}$ ,  $\alpha_1^2 L_{sL}^{(1)}$ , Eq. (38) takes the form

$$P_{rc} = \beta_0 L_{31} L_{sL}^{(1)} / (L_{31} + L_{sL}^{(1)}). \quad (40)$$

If  $L_{31} \gg L_{sL}^{(1)}$ , then the "bottleneck" of the energy transfer from the alternating field to the lattice is the transition of the energy from the spin system to the lattice, and from (40) we have  $P_{rc} = \beta_0 L_{sL}^{(1)}$ .

At  $L_{sL}^{(1)} \gg L_{31}$ , the "bottleneck" is the transition of the energy from the field to the spin system, and  $P_{rc} = \beta_0 L_{31}$ .

To determine the conditions under which formula (40) is applicable, it is more convenient to compare not  $L_{21}$  with  $L_{sL}^{(2)}$ , but the corresponding probabilities

$$W_2 = L_{21} / \langle \mathcal{H}_2^2 \rangle \approx \tau^{-3} / \omega_0^2 \quad (41)$$

and

$$T_{1(2)}^{-1} = L_{sL}^{(2)} / \langle \mathcal{H}_2^2 \rangle \approx T_{1(1)}^{-1}. \quad (42)$$

It is seen from (41) that in ESR the condition  $L_{21} \gg L_{sL}^{(2)}$  is easily satisfied for a field  $H_0 \approx 10^3$  Oe. This circumstance can serve as a simple explanation of the well-known fact that in the case of ESR it is difficult to observe effects connected with the existence of two spin temperatures. In the case of NMR, the condition  $L_{21} \gg L_{sL}^{(2)}$  is satisfied for fields on the order of  $10^3$  Oe at large  $T_1$  (usually at  $T_1 > 10^2$  sec). For objects with shorter  $T_1$  it is possible to attain the condition  $L_{21} \gg L_{sL}^{(2)}$  by decreasing  $H_0$ .

As to the experiments known to me on the role of DDP in ESR and NMR, they were performed under conditions when  $L_{21} \ll L_{sL}^{(2)}$ . In Atsarkin's<sup>[9]</sup> investigation, for example, of the  $Ce^{3+}$  in the  $CaWO_4$  crystal, the condition  $L_{21} \ll L_{sL}^{(2)}$  is satisfied in spite of the very large time  $T_1$  ( $T_1 = 4$  sec at  $1.8^\circ K$ ), since at concentrations  $10^{18}$  cm<sup>3</sup> the  $Ce^{3+}$  ion in  $CaWO_4$  has  $T_2 \approx 5 \times 10^{-6}$  sec. By

increasing the  $Ce^{3+}$  concentration in  $CaWO_4$  it is possible to satisfy the condition  $L_{21} \gg L_{sL}^{(2)}$  for this object.

A transition from the case  $L_{21} > L_{sL}^{(2)}$  to the case  $L_{21} < L_{sL}^{(2)}$  is possible also for a single object by changing its temperature.

#### 4. CONCLUSION

As we have shown in Sec. 2, allowance for the satellite of the  $J(\omega)$  curve at the frequency  $\omega=0$  leads to a Lorentzian dependence of the probability  $W_2$  on  $H_0$  for sufficiently strong fields  $H_0$ , regardless of the shape of the absorption curve. The analysis at the end of Sec. 2 shows that this result does not contradict the available experimental data. It seems of interest to measure the rate of establishment of a single-temperature in the spin system under conditions when the results of Sec. 2 are significant, that is, at values of  $H_0$  exceeding  $H_{1oc}$  by several times. In Sec. 3, using the results of Sec. 2, equations were obtained describing the magnetic resonant absorption of energy of an alternating field, and an explanation was presented of the difficulties that arise in the observation, in ESR, of effects connected with the existence of two spin temperatures. It would be of interest to perform experiments aimed at determining the role of the DDP in magnetic resonance under conditions of the transition from the

case  $L_{21} < L_{sL}^{(2)}$  to the case  $L_{21} > L_{sL}^{(2)}$ . From the theoretical point of view, it may be useful to study the  $J(\omega)$  curve near  $\omega=0$  in order to determine the coefficient  $L_{21}$  more accurately.

The author is grateful to Professor B. I. Kochelaev for useful advice and for a discussion of the results.

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Translated by J. G. Adashko

## Instabilities and characteristics of galvanomagnetic effects in inhomogeneous films subjected to crossed fields

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(Submitted June 3, 1975)

*Zh. Eksp. Teor. Fiz.* 70, 569-577 (February 1976)

A theoretical analysis is made of galvanomagnetic effects in thin semiconducting films with inhomogeneous distributions of the carrier density and mobility across the film thickness, subjected to crossed  $\mathbf{E}$  and  $\mathbf{H}$  fields parallel to the film surface. It is shown theoretically that the galvanomagnetic coefficients of inhomogeneous films are greatly affected by reversal of the sign of  $\mathbf{E} \times \mathbf{H}$ . This nonreciprocity of the coefficients is due to a redistribution of carriers across the film thickness by the Lorentz force. The results are given of an experimental investigation of galvanomagnetic properties of pure two-layer epitaxial films of  $n$ -type GaAs in the temperature range 4.2-300°K. The predicted nonreciprocity of the galvanomagnetic coefficients is observed and a current instability, nonreciprocal in respect of the direction of the Lorentz force, is found. This instability is observed in fields much lower than the Gunn fields in the absence of a falling region in the current-voltage characteristic but in the presence of a falling region in the current-magnetic field characteristic.

PACS numbers: 73.60.Fw

1. A Hall field, which cancels the Lorentz force acting on free carriers, appears in semiconducting films subjected to crossed electric and magnetic fields parallel to the film surface. The source of the Hall field are free carriers deflected by the Lorentz force to one of the surfaces of the film. In the case of thick films with high electron or hole densities, only a slight redistribution of charged particles is needed to cancel the Lorentz force by the Hall field. In pure and thin films subjected

to strong  $\mathbf{E}$  and  $\mathbf{H}$  fields parallel to the film surface a strong redistribution of carriers across the thickness is needed to create the necessary Hall field and, therefore, it is interesting to determine how this redistribution affects the galvanomagnetic properties of semiconducting films.

We must bear in mind that the Hall field, which appears in a film because of a redistribution of impuri-