

on curves 1 and 2 of Fig. 2 in a field 11 kOe ($\alpha \approx 2$) is equal to 14. The theoretical ratio of the amplitude of the doppleron oscillations to the GKO amplitude in the same field is 9. We do not compare the results on the GKO in minus polarization and due to holes. The reason is that the theory constructed above does not take into account non-local effects connected with the holes.

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Cross relaxation between the Zeeman and spin-spin degrees of freedom in a solid

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A theory is presented of the establishment of a single spin temperature under the influence of a nonsecular dipole-dipole interaction \mathcal{H}''_d , in higher orders of perturbation theory. It is shown that when the cross relaxation processes are considered it is necessary to exclude in each succeeding approximation the secular contribution from the perturbation \mathcal{H}''_d and to redefine correspondingly both the Zeeman subsystem and the subsystem of the spin-spin interactions. It is pointed out that the ideas presently advanced in the literature, concerning the unification of the Zeeman subsystem with \mathcal{H}''_d , are inconsistent.

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The dipole-pool concept, advanced by Provotorov,^[1] has permitted considerable progress to be made in research on magnetic resonance.^[2] The gist of this concept is that in strong constant magnetic fields $H_0 \gg H_L$ (H_L is the field due to the dipole-dipole (dd) interaction of the given spin with the environment) the populations of the aggregates of the levels which result from the lifting of the degeneracy of the Zeeman levels of the spin sys-

tem by the secular part \mathcal{H}'_d of the dd interaction are determined by a temperature β_d^{-1} that is in general different from the Zeeman temperature β_z^{-1} . It is the interaction \mathcal{H}'_d , regarded as a thermodynamic subsystem with temperature β_d^{-1} , which is in fact called the dipole pool.

The Provotorov two-temperature model has a clear physical foundation. \mathcal{H}'_d conserves the Zeeman energy

\mathcal{H}_z , so that the flip-flop processes induced by \mathcal{H}'_d proceed much more rapidly than the processes connected with the nonsecular part \mathcal{H}''_d of the dd interaction. Therefore, a quasi-equilibrium state is produced in the spin system after the lapse of the spin-spin relaxation time T_2 induced by the interaction \mathcal{H}'_d , and is characterized by two quasi-integrals of motion, \mathcal{H}_z and \mathcal{H}'_d . It becomes possible therefore to describe irreversible processes macroscopically with the aid of the two reciprocal temperatures β_z and β_d . In this case the "slow" interaction

$$\mathcal{H}''_d = \sum_{m=-2,1,2} \mathcal{H}''^{(m)}_d$$

is responsible for the thermal mixing of the Zeeman and dipole pools.¹⁾

It must be borne in mind, however, that the described model is valid only in the lowest order of perturbation theory in the dd interaction \mathcal{H}_d . If we are interested in the next-order approximation,^[4] then the secular part of the dipole-dipole interaction takes the form²⁾ $\mathcal{H}'_d + \mathcal{H}''_{ds}$, where

$$\mathcal{H}''_{ds} = \sum_{m \neq 0} \frac{1}{2m\omega_0} [\mathcal{H}''^{(m)}_d, \mathcal{H}''^{(-m)}_d].$$

The operator \mathcal{H}''_{ds} corresponds to the correction that must be introduced into the Zeeman energy in second-order perturbation theory in \mathcal{H}_d .

It is important to note that \mathcal{H}''_{ds} gives a non-zero first moment of the absorption line shape $f(\omega)$:

$$M_1 = \int_{-\infty}^{\infty} \omega f(\omega) d\omega = \frac{\omega_0}{2} \frac{\langle (\mathcal{H}''_d)^2 \rangle}{\langle \mathcal{H}^2_z \rangle}, \quad \langle \dots \rangle = \frac{\text{Sp}(\dots)}{\text{Sp} 1},$$

and consequently causes a homogeneous shift of the Zeeman frequency ω_0 . Therefore the corresponding contribution must be combined with \mathcal{H}_z , defining a "shifted" Zeeman pool $\tilde{\mathcal{H}}_z = \mathcal{H}_z + M_1 S_z$. Then the modified secular part of the dd interaction, which broadens the line and gives rise to processes that proceed with conservation of $\tilde{\mathcal{H}}_z$, takes the form³⁾

$$\tilde{\mathcal{H}}_z = \mathcal{H}'_d + \mathcal{H}''_{ds} - M_1 S_z.$$

Under certain conditions and when $\mathcal{H}'_d \approx 0$, it is precisely $\mathcal{H}''_{ds} - M_1 S_z$ which is responsible for the width of the magnetic resonance^[5] and for the spin diffusion,^[6] and also leads to establishment of internal equilibrium in each of the spin subsystems.

The connection between $\tilde{\mathcal{H}}_z$ and $\tilde{\mathcal{H}}_d$ is effected by the "truly" nonsecular interaction $\mathcal{H}''_{d's} = \mathcal{H}''_d - \mathcal{H}''_{ds}$. In this situation, the local-equilibrium statistical operator (SO) must be written in the high-temperature approximation in the following form:

$$\rho_q = \frac{1}{\text{Sp} 1} \{1 - \beta_z \tilde{\mathcal{H}}_z - \beta_d \tilde{\mathcal{H}}_d\}. \quad (1)$$

In accordance with the general definition of the subsys-

tems,^[7] $\mathcal{H}''_{d's}$ must not enter explicitly in $\tilde{\rho}_q$.

As noted in^[4] and as follows from simple calculations, the average energy of the spin system, in second order in the small parameter H_L/H_0 , obtained with the aid of $\tilde{\rho}_q$, coincides with the result that follows from the use of the SO:

$$\rho_q = \frac{1}{\text{Sp} 1} \{1 - \beta_z (\mathcal{H}_z + \mathcal{H}''_d) - \beta_d \mathcal{H}'_d\}. \quad (2)$$

We note that in SO that practically coincides with (2) was introduced by Goldman,^[3] who started from the analogy between the problem considered here and the problem of describing a spin system in the presence of a strong saturating field in a rotating coordinate system. The separated subsystems are $Z^* = \mathcal{H}_z + \mathcal{H}''_d$ and $\mathcal{H}'_{d^*} = \mathcal{H}'_d - V$, and the interaction V between Z^* and \mathcal{H}'_{d^*} is chosen from the commutation condition $[Z^*, \mathcal{H}'_{d^*}] = 0$ (the explicit form of V is written out in^[3] in the lowest-order approximation). The local-equilibrium SO is written in the form

$$\rho_{q^*} = \frac{1}{\text{Sp} 1} \{1 - \beta_z Z^* - \beta_d \mathcal{H}'_{d^*}\}. \quad (3)$$

The main difference between the SO (2) or (3) and the SO (1) is that \mathcal{H}_z and \mathcal{H}''_d are combined into a single subsystem. Therefore the physical justification given in^[3] for formula (3) implies in fact this unification during the initial stage of the abbreviation of the description (it is difficult to agree with this procedure if one starts from energy considerations).

At the same time, the form of the local-equilibrium SO determines uniquely the non-equilibrium SO (NSO),^[8] i. e., the kinetics of the processes. If the SO (2) and (3) are formally equivalent to (1) not only in the sense that the average energies of the spin system coincide in the second-order approximation in H_L/H_0 , but describe in fact the macroscopic state of the system and its time evolution, then all the operators (1)–(3) should lead to the same values of the kinetic coefficients in second-order perturbation theory. Let us examine this question in greater detail.

We consider the spin system, which is insulated from the lattice, of a solid in a constant magnetic field $H_0 \parallel z$. We write down the Hamiltonian of the system in the form

$$\mathcal{H} = \mathcal{H}_z + \mathcal{H}'_d + \mathcal{H}''_d = \tilde{\mathcal{H}}_z + \tilde{\mathcal{H}}_d + \tilde{\mathcal{H}}''_{d's}, \quad \mathcal{H}_z = \omega_0 S_z, \quad (4)$$

and assume that $H_0 \gg H_L$.

As indicated above, to describe the irreversible processes in first-order in H_L/H_0 we can use Provotorov's usual scheme, separating the subsystems \mathcal{H}_z and \mathcal{H}'_d with reciprocal temperatures β_z and β_d , and regarding \mathcal{H}''_d as a small perturbation. With the aid of the local-equilibrium SO

$$\rho_{q'} = \frac{1}{\text{Sp} 1} \{1 - \beta_z \mathcal{H}_z - \beta_d \mathcal{H}'_d\} \quad (5)$$

(1) coincides with (5) (in this approximation) we obtain the system

$$\frac{d\beta_z}{dt} = -\frac{\beta_z - \beta_d}{T_{zd}'} = -\frac{\langle \mathcal{H}_d'^2 \rangle}{\langle \mathcal{H}_z'^2 \rangle} \frac{d\beta_d}{dt}, \quad (6)$$

where

$$\frac{1}{T_{zd}'} = -\frac{1}{\langle \mathcal{H}_z'^2 \rangle} \int_{-\infty}^0 e^{t'} \langle [\mathcal{H}_d', \mathcal{H}_d''] \cdot [\mathcal{H}_d', \mathcal{H}_d'']^0(t) \rangle dt \quad (7)$$

and the dependence on t in the correlator under the integral sign means the representation of the interaction with the Hamiltonian $\mathcal{H}_0 = \mathcal{H}_z + \mathcal{H}_d$. Simple calculations yield

$$\frac{1}{T_{zd}'} = \frac{\pi}{\langle S_z'^2 \rangle} \sum_{m \neq 0} \langle \mathcal{H}_d''^{(m)} \mathcal{H}_d''^{(-m)} \rangle f_m(\omega_0), \quad (8)$$

where $f_m(\omega)$ is the Fourier transform of the correlator

$$f_m(t) = \frac{\langle \mathcal{H}_d''^{(m)} \exp(i\mathcal{H}_d' t) \mathcal{H}_d''^{(-m)} \exp(-i\mathcal{H}_d' t) \rangle}{\langle \mathcal{H}_d''^{(m)} \mathcal{H}_d''^{(-m)} \rangle}. \quad (9)$$

It is easy to verify that in first order in H_L/H_0 the same result is obtained by using an SO in which an arbitrary reciprocal temperature is assigned to the interaction \mathcal{H}_d'' (in particular, the temperature β_z^{-1} , just as in (2)), so that from the point of view of the theory of irreversible processes, within the framework of our approximation, the unification of the perturbation \mathcal{H}_d'' with any subsystem whatever is meaningless.

Analogously, in case (3), we obtain for the rate of thermal mixing between Z^* and $\mathcal{H}_d'^*$, in the lowest order in V , the expression

$$\frac{1}{T_{zd}'} = -\frac{1}{\langle Z'^2 \rangle} \int_{-\infty}^0 e^{t'} \langle [Z', V] \cdot [Z', V]^0(t) \rangle dt.$$

Inasmuch as $[Z^*, \mathcal{H}_d'^*] = 0$, we have

$$[Z', V] = [Z', V + \mathcal{H}_d'] = [Z', \mathcal{H}_d'] = [\mathcal{H}_d'', \mathcal{H}_d'], \quad (10)$$

so that $T_{zd}^* = T_{zd}'$ in first order in H_L/H_0 . Thus, Goldman's analysis^[3] in the lowest-order approximation likewise contains nothing new from the physical point of view.

The situation is entirely different if higher-order approximations are used. We confine ourselves for simplicity to second order in H_L/H_0 . Using the reasoning presented above, we construct an NSO^[8] on the basis of (1) in the form

$$\rho = e \int_{-\infty}^0 e^{t'} e^{i t' H_0} \rho_0(t+t') e^{-i t' H_0} dt' = \frac{1}{\text{Sp } 1} \left\{ 1 - \beta_z \mathcal{H}_z - \beta_d \mathcal{H}_d + \int_{-\infty}^0 e^{t''} \frac{d}{dt'} [\beta_z(t+t') \mathcal{H}_z(t') + \beta_d(t+t') \mathcal{H}_d(t')] dt' \right\}, \quad (11)$$

where the time dependence of the operators is defined in the Heisenberg approximation with total Hamiltonian (4).

Averaging the equations of motion

$$\frac{d\tilde{\mathcal{H}}_{z,d}(t)}{dt} = \frac{1}{i} [\tilde{\mathcal{H}}_{z,d}(t), \mathcal{H}_{ds}'(t)]$$

with the aid of the NSO (11), we can obtain a system (6)

with a mixing rate

$$\frac{1}{T_{zd}} = -\frac{1}{\langle \mathcal{H}_z'^2 \rangle} \int_{-\infty}^0 e^{t'} \langle [\tilde{\mathcal{H}}_z, \mathcal{H}_{ds}'] \cdot [\tilde{\mathcal{H}}_z, \mathcal{H}_{ds}']^0(t) \rangle dt. \quad (12)$$

If we start from (2), then we obtain for the mixing rate the expression^[9]

$$\frac{1}{T_{zd}} = -\frac{1}{\langle \mathcal{H}_z'^2 \rangle} \int_{-\infty}^0 e^{t'} \langle [\mathcal{H}_d', \mathcal{H}_d''] \cdot [\mathcal{H}_d', \mathcal{H}_d'']^0(t) \rangle dt, \quad (13)$$

in which, just as in (12), the time dependence is determined by the Hamiltonian^[4] (4).

Calculating \tilde{T}_{zd}^{-1} in second-order in H_L/H_0 , we obtain after straightforward but laborious calculations:

$$\frac{1}{T_{zd}} = \frac{1}{T_{zd}'} - \frac{\pi}{4\langle \mathcal{H}_z'^2 \rangle} \sum_{\substack{m \neq 0, m' \neq 0 \\ (m', m-m)}} \frac{1}{mm'} C_{mm'}(\omega_0(m+m')), \quad (14)$$

where $C_{mm'}(\omega)$ is the Fourier transform of the correlator

$$C_{mm'}(t) = \langle [\mathcal{H}_d', [\mathcal{H}_d''^{(m)}, \mathcal{H}_d''^{(m')}] \cdot [\mathcal{H}_d', [\mathcal{H}_d''^{(m)}, \mathcal{H}_d''^{(m')}]]^0(t) \rangle. \quad (15)$$

The dependence on t in (15) is brought about by the interaction \mathcal{H}_d' .

Calculation of the mixing rate (13)^[9] in second-order approximation leads to formula (14), the only difference being that an additional term with $m' = -m$ appears and can be reduced to the expression

$$-\frac{\pi}{4\langle \mathcal{H}_z'^2 \rangle} \sum_{m \neq 0} \frac{C_{m,-m}(0)}{m^2} = -\frac{\pi}{\langle S_z'^2 \rangle} \langle [\mathcal{H}_d', \mathcal{H}_{ds}'] \cdot [\mathcal{H}_d', \mathcal{H}_{ds}']^0(t) \rangle_{\omega=0}. \quad (16)$$

We note that, by virtue of (10), precisely the same result is obtained also in Goldman's scheme.^{[3] 5)}

Formula (16) corresponds to a satellite at zero frequency.^[9] Thus, in the approaches used in^[9,3], in contrast to formula (14), the mixing rate contains a contribution that depends on the constant magnetic field like $T_{zd}^{-1} \sim H_0^{-2}$, independently of the form of the correlators $C_{mm'}(\omega)$ and $f_m(\omega)$. Since the correlation functions usually decrease with increasing H_0 more rapidly than the power-law function H_0^{-2} , Skrebnev^[9] draws the physically important conclusion that the mixing of the Zeeman and dipole pools in strong fields is due mainly to just this term, i.e., is a rather rapid process.

It is obvious from the foregoing that the appearance of a satellite at zero frequency is the consequence of the unification of \mathcal{H}_z and \mathcal{H}_d' into a single subsystem and, as follows from (16), is in fact equivalent to application of perturbation theory to the secular interaction \mathcal{H}_{ds}' . Although this does not influence substantially the average spin-system energy, it does lead to incorrect expressions for the kinetic coefficients in the higher approximations.

It is useful to emphasize here the connection between the result (16) and the appearance of terms proportional to t^2 (these are the so-called secular terms in the usual perturbation-theory expansion.^[10] The elimination of \mathcal{H}_{ds}' from \mathcal{H}_d' and exact allowance for \mathcal{H}_{ds}' together with

\mathcal{H}'_d , as is done when the correct result (14) is obtained, corresponds to elimination of the secular contributions from the perturbation-theory series.^[10]

Thus, it can be stated that the reasons why the result $T_{dd}^{-1} \sim H_0^{-2}$ and all its consequences^[9] are in error are, in final analysis, both the unification of \mathcal{H}_z and \mathcal{H}''_d and the incorrect separation of the secular part of the dd interaction and the formal application of perturbation theory to \mathcal{H}''_d . The interaction \mathcal{H}''_d contains in this approximation an "undesirable" secular contribution \mathcal{H}''_{ds} , which must be eliminated from \mathcal{H}''_d .

The problem discussed above can be understood also on the basis of the general principles of statistical mechanics of irreversible processes,^[11,13] according to which the macroscopic coordinates of the system must be secular variables, such as \mathcal{H}_z and \mathcal{H}_d are in our analysis. The formal choice of the subsystems in^[3,9], as shown in the present paper, can lead to incorrect physical results.

It is clear from all the foregoing that the unification of \mathcal{H}_z and \mathcal{H}''_d in the lowest order of perturbation theory does not lead to any new physical results whatever in comparison with the usual separation of the Zeeman and dipole subsystems in accordance with Provotorov's scheme. On the other hand, in the higher orders in H_L/H_0 , this unification leads to erroneous result and must be rejected. The consistent method of taking the higher-order approximations into account can be understood from the foregoing reasoning. In each next order in H_L/H_0 , starting already with the second, it is necessary to eliminate from the interaction \mathcal{H}''_d , which brings about an equilibrium between the Zeeman and dipole pools, the secular terms of the corresponding order, and to include their first moment in \mathcal{H}_z , while the remainder is unified with \mathcal{H}'_d . It is then clear that regardless of the temperature that is ascribed to the perturbation $\mathcal{H}''_{ds} = \mathcal{H}''_d - \mathcal{H}''_{ds}$, "secular" processes that do not alter \mathcal{H}_z will be excluded in the course of the energy exchange between the "correct" Zeeman subsystem \mathcal{H}_z and the "correct" dipole pool \mathcal{H}_d in any order of perturbation theory. For this reason, the "correct" correlation functions of the type contained in (12) will not contain the zero-frequency satellites that result from the pres-

ence of x secular terms in the perturbation.

We note in conclusion that the arguments advanced here are important also in the analysis of cross relaxation and thermal mixing of the Zeeman and dipole pools in a rotating coordinate system.

- ¹The explicit forms of the operators \mathcal{H}'_d and $\mathcal{H}''_d^{(m)}$ can be found, for example, in^[3].
- ²Here and below we use the results of^[4] in operator form.
- ³We note that \mathcal{H}''_{ds} , in contrast to \mathcal{H}'_d , describes four-spin processes.
- ⁴We have left out from (12) and (13) terms that are of no importance in the analysis and are due to the presence of the derivatives $(d/dt')\beta_{z,d}(t+t')$ in (11).
- ⁵Goldman himself confined his analysis to the lowest order in the calculation of the kinetic coefficients. We note, incidentally, that the results of Chap. 6 of^[3] can be easily derived on the basis of (1), without resorting at all to the unification of \mathcal{H}_z with \mathcal{H}''_d .
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