the electronic states with changing number of neighboring iron atoms. In addition, μ^i(Mn) can be greatly influenced by the delocalized spin density, the spatial distribution of which has a complicated character and depends to a considerable degree on the type and the distribution of the neighboring atoms.

The solution of the system (1) has made it possible to determine the coefficients of the hyperfine interaction in the relation

$$H^i(\mathrm{Mn}) = a\mu^i(\mathrm{Mn}) + b\bar{\mu},$$

which turn out to be $a = -40$ kG/μ_B and $b = -100$ kG/μ_B. The values of these coefficients point to an appreciable degree of delocalization of the spin density in the alloys $(Fe_{1-x}Mn_x)_3Al$.

For samples 3 and 4 with maximum manganese contents, we determined also the relative numbers of manganese atoms with antiferromagnetic orientation μ(Mn). It turned out that for sample 3 about 50% of the manganese atoms situated at the Fe_{II} sites have antiferromagnetic orientation. For sample 4, this value is 60%. This conclusion is confirmed also by neutron-diffraction data, which have established the presence of manganese-enriched segregations in samples 3 and 4. The probability of flipping of μ(Mn) in such segregations, by virtue of the negative exchange interaction between the neighboring manganese atoms, can be quite high.

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Contribution to spin-spin cross relaxation theory

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Evolution of the spin system of a solid paramagnet after the sudden introduction of frequency detuning Δ between the subsystems involved in cross relaxation (CR) is considered. The calculation is carried out by the density-matrix technique using projection operators and is applied to the general case of nonequidistant EPR and NMR spectra. It is shown that after a time of the order of ω_d^{-1}(ω_d is the local field frequency) the energy of the nonsecular part of the spin-spin interactions, which is responsible for the CR, is mixed with a part of the Zeeman energy, thus forming a quasiequilibrum "difference" pool. At $|\Delta| \gg \omega_d$ the process resembles damped oscillations and can be related via a Fourier transformation to the function between the CR probability and Δ. An experiment is proposed by which the predicted effects may be observed and employed for investigation of spin-spin interaction in solids.

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1. INTRODUCTION

The study of the processes of cross relaxation (CR) in parametric spin systems has already been the subject of hundreds of theoretical and experimental studies (see, e.g., [1–5]), but many important aspects of this phenomenon still remain unclear and continue to attract attention of researchers. One such unsolved problem is connected with the behavior of that of the "nonsecular" terms of the Hamiltonian of the dipole-dipole (DD) interactions,[1)] which induces mutual spin flips I and S with close resonant frequencies ω_I and ω_S. For pure spin magnetism, it takes the form[1]

$$\hat{\mathscr{H}}_{CR} = \sum b_{ij}(\hat{I}_i^+\hat{S}_j^- + \hat{I}_i^-\hat{S}_j^+).$$

Although with respect to its specific heat this term can be comparable with the secular part $\hat{\mathscr{H}}_d'$ of the DD interactions, its contribution to the total energy balance is not taken into account in the spin-temperature theory of CR.[2,3] This contradiction manifests itself clearly on going to the "resonant CR," corresponding to the exact agreement of the frequencies ω_I and ω_S, while CR with large detuning $\Delta = \omega_I - \omega_S \gg \omega_d$, where ω_d is the average frequency of the dipole-dipole pool (DDP). Whereas in the former case the term $\hat{\mathscr{H}}_{CR}$ becomes secular and is included in the DDP, in the latter case its specific heat is neglected, so that the increase of Δ seems to be connected with the "vanishing" of the energy stored in $\hat{\mathscr{H}}_{CR}$. This seems particularly paradoxical if the DDP is strongly cooled beforehand by any one of the known methods.

In the recent literature two trends are noted in the solution of this problem, which turned out to be closely related with the fundamental problems of establishment of quasi-equilibrium (the formation of the temperatures) in spin systems. Thus, in[6] $\hat{\mathscr{H}}_{CR}$ is classified as a separate pool with its own temperature, as a result of which the entire CR picture is substantially altered. The other trend was proposed in[3,4], where an analogy is made between the situation in question and the behavior of a simple system of spins of the same sort when an external magnetic field H_0 is turned on rapidly. As shown in[4,7,8], in this case the energy of the nonsecular part $\hat{\mathscr{H}}_d''$ of the DD interaction is transferred during the time of the transverse relaxation $\tau_2 \sim \omega_d^{-1}$ to the Zeeman subsystem $\hat{\mathscr{H}}_z$, and this leads to the experimentally observable change of the longitudinal magnetization, and is interpreted in[3,4,8] as formation of a single energy pool $\hat{\mathscr{H}}_z + \hat{\mathscr{H}}_d''$.

In this paper we consider the behavior of a system in which CR takes place with the detuning Δ turned on rapidly. It will be shown that the results of this analysis confirm the usefulness of the approach indicated in[3,4], and the formation of new quasi-equilibrium energy pool is the result of energy transfer from $\hat{\mathscr{H}}_{CR}$ to the "difference" part of the Zeeman subsystem, while the experimental observation of the corresponding fast transient process can serve as a new effective method of studying DD interactions in solids.

2. INITIAL HAMILTONIAN

We consider a solid paramagnet placed in a constant magnetic field H_0, in which the DD interactions are assumed to be strong in comparison with the spin-lattice interactions, and the role of the lattice is henceforth disregarded. The CR process is frequently considered in systems with two sorts of spins, I and S, that experience pure magnetic interactions, with the Hamiltonian of the system taking the form

$$\hat{\mathscr{H}} = \omega_I \hat{I}_z + \omega_S \hat{S}_z + \hat{\mathscr{H}}_d' + \hat{\mathscr{H}}_d'' + \hat{\mathscr{H}}_{CR}, \tag{1}$$

where

$$\hat{I}_z = \sum_i \hat{I}_z^i, \quad \hat{S}_z = \sum_i \hat{S}_z^i,$$

$\hat{\mathscr{H}}_d'$ and $\hat{\mathscr{H}}_d'' + \hat{\mathscr{H}}_{CR}$ are respectively the secular and nonsecular parts of the DD interactions.

The first two terms of (1) can also be written in the form

$$\omega_0 (\hat{I}_z + \hat{S}_z) + \Delta (c_S \hat{I}_z - c_I \hat{S}_z) \equiv \hat{\mathscr{H}}_\Sigma + \hat{\mathscr{H}}_\Delta,$$

where $\omega_0 = c_I \omega_I + c_S \omega_S$, and c_I and c_S are the relative specific heats of the subsystems I and S; $c_I + c_S = 1$. This notation is useful under CR conditions, when only the "difference" or Δ subsystem $\hat{\mathscr{H}}_\Delta$ changes, whereas the energy of the "Σ subsystem" $\hat{\mathscr{H}}_\Sigma$ is an integral of the motion and can be completely disregarded.[3,9] Systems with pure spin magnetism are described in the I_z and S_z representation, a procedure that offers many advantages. However, the "resonant CR" ($\Delta = 0$) which is of interest to us is realized here only in the trivial case $\omega_I = \omega_S = 0$.

In practice, CR processes are more frequently encountered in more complicated systems, the Hamiltonian of which includes (for a spin > 1/2) the energy of the electron paramagnetic center in the intracrystalline field, or else the energy of the nuclear quadrupole interaction. In this case the role of the subsystems I and S can, in particular, be assumed by different transitions in non-equidistant multilevel energy spectra. The main differences between such systems and "pure" spin systems are the following: 1) resonant CR ($\Delta = 0$) is possible at $\omega_I \gg \omega_d$ and $\omega_S \gg \omega_d$, the detuning Δ being a function of the magnitude and direction of $\mathbf{H}_0$, and is thus easily controlled in experiment; 2) the particle energies are not purely magnetic, so that it is necessary to redefine the subsystems I and S as well as the secular and nonsecular parts of the DD interactions; 3) by virtue of the selection rules (see, e.g.),[1] the resonant-CR time W_{CR}^{-1} can be much longer than τ_2, so that even at $\Delta = 0$ it is possible to describe the subsystems I and S separately with the aid of the corresponding temperatures.

It is more convenient to carry out the description in this case in the energy representation. We define a projection operator P_α^i (see, e.g.[10]) acting only on the wave functions of the i-th spin ψ_β^i, calculated neglecting the DD interactions: $\hat{P}_\alpha^i \psi_\beta^i = \delta_{\alpha\beta} \psi_\beta^i$, where $\delta_{\alpha\beta}$ is the Kronecker symbol and the Greek indices number the energy levels of the system. We shall henceforth assume that all the levels are nondegenerate, and then the Hamiltonian of the system without allowance for the DD interactions takes the form

$$\hat{\mathscr{H}}^{(0)} = \frac{1}{\hbar} \sum_{\alpha'=1}^{R} \sum_{i=1}^{N} E_{\alpha'} \hat{P}_{\alpha'}^i = \frac{1}{\hbar} \sum_{\alpha'=1}^{R} E_{\alpha'} \hat{N}_{\alpha'}, \tag{2}$$

where $E_{\alpha'}$ is the energy of the α'-th level of one particle, R is the total number of levels, N is the number of particles, and $\hat{N}_{\alpha'}$ are the level-population operators (see, e.g.,[4]).

Assume now that for any two transitions, say $\alpha\beta$ and $\gamma\delta$, of the multilevel spectrum there takes place an effective CR, which calls for satisfaction of the condition $\omega_I \approx \omega_S$, where

$$\omega_S \equiv \omega_{\alpha\beta} = (E_\beta - E_\alpha)/\hbar, \quad \omega_I \equiv \omega_{\gamma\delta} = (E_\delta - E_\gamma)/\hbar.$$

To consider the CR between the chosen transitions we replace the operators $\hat{N}_\alpha$, $\hat{N}_\beta$, $\hat{N}_\gamma$ and $\hat{N}_\delta$ by their linear combinations $(\hat{N}_\alpha + \hat{N}_\beta)/2$, $(\hat{N}_\gamma + \hat{N}_\delta)/2$, $\hat{N}_S = (\hat{N}_\beta - \hat{N}_\alpha)/2$, $\hat{N}_I = (\hat{N}_\delta - \hat{N}_\gamma)/2$. The first two operators are integrals of the motion in the CR process, while $\hat{N}_I$ and $\hat{N}_S$ are in effect analogs of $\hat{I}_z$ and $\hat{S}_z$. The operator $\hat{N}_\Sigma = \hat{N}_I + \hat{N}_S$, in analogy with $\hat{I}_z + \hat{S}_z$ in the pure spin case, is one more integral of the motion, while the operator $N_\Delta = (\hat{N}_I - \hat{N}_S)/2$ corresponds to the Δ subsystem. The Hamiltonian (2) takes the following form:

$$\hat{\mathscr{H}}^{(0)} = \frac{1}{\hbar} \sum{}' E_{\alpha'} \hat{N}_{\alpha'} + \frac{E_\alpha + E_\beta}{2\hbar} (\hat{N}_\alpha + \hat{N}_\beta) + \frac{E_\gamma + E_\delta}{2\hbar} (\hat{N}_\gamma + \hat{N}_\delta) + \omega_0 \hat{N}_\Sigma + \Delta \hat{N}_\Delta, \tag{3}$$

where the prime at the summation sign denotes summation over all the indices except α, β, γ and δ; $\omega_0 = (\omega_I$

$+\omega_S)/2$, $\Delta=\omega_I-\omega_S$. The last term in (3) corresponds to the difference Hamiltonian

$$\hat{\mathscr{H}}_\Delta\equiv\Delta\hat{N}_\Delta={}^1/_4\Delta[(\hat{N}_\delta-\hat{N}_\gamma)-(\hat{N}_\beta-\hat{N}_\alpha)].$$

We note that $\omega_{\gamma\delta}-\omega_{\alpha\beta}=\omega_{\beta\delta}-\omega_{\alpha\gamma}$, and the same expression for $\hat{\mathscr{H}}_\Delta$ is obtained also when another pair of transitions, $\alpha\gamma$ and $\beta\delta$, is considered. With the aid of the operators $\hat{P}^i_\alpha$ we can separate the secular part of the DD interaction $\hat{\mathscr{H}}'_d([\hat{\mathscr{H}}^{(0)}, \hat{\mathscr{H}}'_d]=0)$:

$$\hat{\mathscr{H}}_d'=\sum_{i>j}^{N}\Big[\sum_{\alpha',\beta'=1}^{R}\hat{P}^i_{\alpha'}\hat{P}^j_{\beta'}\hat{\mathscr{H}}_d^{ij}\hat{P}^i_{\alpha'}\hat{P}^j_{\beta'}+\sum_{\alpha'\neq\beta'}^{R}\hat{P}^i_{\alpha'}\hat{P}^j_{\beta'}\hat{\mathscr{H}}_d^{ij}\hat{P}^i_{\beta'}\hat{P}^j_{\alpha'}\Big],\quad (4)$$

where $\hat{\mathscr{H}}^{ij}_d$ is the Hamiltonian of the DD interaction between the spins i and j; the resonant frequencies of all the transitions are assumed here to be different. We can separate analogously also the part $\hat{\mathscr{H}}_{\rm CR}$ responsible for the CR in the case under consideration. Obviously, in this case it is necessary to take into account to the same degree the CR between the transitions $\alpha\beta$, $\gamma\delta$ and $\alpha\gamma$, $\beta\delta$:

$$\hat{\mathscr{H}}_{CR}=\sum_{i>j}^{N}\sum_{\alpha'\beta'\gamma'\delta'}{}'\hat{P}_{\alpha'}{}^i\hat{P}_{\beta'}{}^j\hat{\mathscr{H}}_d{}^{ij}\hat{P}_{\gamma'}{}^i\hat{P}_{\delta'}{}^j.\quad (5)$$

The prime at the summation sign shows that it is necessary to take only those α', β', γ' and δ', to which the terms responsible for the CR correspond (their total number is 8).

In the case of CR between two transitions having a common level, for example $\alpha\beta$ and $\beta\gamma$ ($\omega_{\alpha\beta}\approx\omega_{\beta\gamma}$; $\hbar\omega_{\alpha\beta}=E_\beta-E_\alpha$, $\hbar\omega_{\beta\gamma}=E_\gamma-E_\beta$), the operators $\hat{N}_I=(\hat{N}_\gamma-\hat{N}_\beta)/2$ and $\hat{N}_S=(\hat{N}_\beta-\hat{N}_\alpha)/2$ are no longer linearly-independent, but it is nevertheless meaningful also in this case to introduce the operators $\hat{N}_\Sigma$ and $\hat{N}_\Delta$. Then, for example,

$$\hat{\mathscr{H}}_\Delta=\Delta\hat{N}_\Delta=\frac{\Delta}{6}[(\hat{N}_\gamma-\hat{N}_\beta)-(\hat{N}_\beta-\hat{N}_\alpha)],$$

where $\Delta=\omega_{\beta\gamma}-\omega_{\alpha\beta}$. Expression (4) remains in force, and for $\hat{\mathscr{H}}_{\rm CR}$ we can write a formula similar to (5).

Both parts of the Hamiltonian and density matrix which correspond to integrals of motion can be omitted. In addition, we consider the case when the frequencies of all the transitions are much larger than ω_d, and therefore the term $\hat{\mathscr{H}}''_d$ is inessential. Thus, the initial Hamiltonian in both cases takes the form

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}_\Delta+\hat{\mathscr{H}}_d'+\hat{\mathscr{H}}_{CR}.\quad (6)$$

The described two variants of the CR in multilevel spin systems are the simplest and most typical. With the aid of projection operators it is easy to introduce the Hamiltonians $\hat{\mathscr{H}}_\Delta$, $\hat{\mathscr{H}}'_d$ and $\hat{\mathscr{H}}_{\rm CR}$ also in other cases: in the presence of degeneracy, for other sorts of spins, etc.

3. FIRST STAGE OF THE TRANSIENT PROCESS: ESTABLISHMENT OF QUASI-EQUILIBRIUM AND FORMATION OF SPIN TEMPERATURES

Assume that up to a certain instant $t=0$ the frequencies of any two transitions of the spectrum are exactly equal and $\hat{\mathscr{H}}_{\rm CR}$ is included in the secular part of the DD interactions. Assume furthermore that by that instant of time the differences of the populations of the considered transitions have become equalized, so that the difference part of the density matrix, corresponding to the atom $\hat{\mathscr{H}}_\Delta$ is equal to zero. Then in the high-temperature approximation the density matrix takes the form

$$\hat{\rho}_0=C[1-\beta_d(\hat{\mathscr{H}}_d'+\hat{\mathscr{H}}_{CR})],$$

where β_d^{-1} is the DDP temperature.

Assume that at the instant $t=0$ a detuning Δ has suddenly been produced between the frequencies of the considered transitions (on account of a jump in the magnitude or direction of $\mathbf{H}_0$), and the Hamiltonian has acquired the form (6). As we shall show, the relaxation of the system to the new equilibrium state will proceed in two stages: the first terminates within a time on the order of ω_d^{-1}, and the second within a time $W_{\rm CR}^{-1}$, which is usually much longer than ω_d^{-1}. In this section we are interested only in the first stage of the transient process.

The condition $|\Delta|\ll\omega_{\alpha,\beta}$ enables us to neglect the change in the eigenfunctions Ψ^i_α, and consequently also the operators $\hat{P}^i_\alpha$, $\hat{N}_\Delta$, $\hat{\mathscr{H}}'_d$ and $\hat{\mathscr{H}}_{\rm CR}$ when the detuning is introduced. Taking this into account, we change over to the interaction representation, in which each new operator $\hat{A}^*$ is connected with the corresponding operator $\hat{A}$ by the formula

$$\hat{A}^*=\exp[i(\hat{\mathscr{H}}_\Delta+\hat{\mathscr{H}}_d')t]\hat{A}\exp[-i(\hat{\mathscr{H}}_\Delta+\hat{\mathscr{H}}_d')t].$$

In this representation, the evolution of the density matrix is described by the equation

$$\partial\hat{\rho}^*(t)/\partial t=-i[\hat{\mathscr{H}}_{CR}{}^*(t),\hat{\rho}^*(t)],$$

and for the mean value of the energy $\langle\hat{\mathscr{H}}'^*_d+\hat{\mathscr{H}}^*_{\rm CR}\rangle$ we have

$$\langle\hat{\mathscr{H}}_d'^*+\hat{\mathscr{H}}_{CR}^*\rangle(t)=\mathrm{Sp}\,\{[\hat{\mathscr{H}}_d'^*(t)+\hat{\mathscr{H}}_{CR}^*(t)]\hat{\rho}^*(t)\}.\quad (7)$$

We confine ourselves for the time being to the case $|\Delta|\gg\omega_d$, and then we can use for the intervals $t\ll W_{\rm CR}^{-1}$ the approximation $\hat{\rho}^*(t)\approx\hat{\rho}^*(0)=\hat{\rho}_0$, corresponding to inclusion in the expression for $\hat{\rho}(t)$ of small terms up to order ω_d/Δ inclusive.

It is easy to obtain the following commutation rules:

$$[\hat{\mathscr{H}}_\Delta,\hat{\mathscr{H}}_{CR}]=\Delta\hat{\tilde{\mathscr{H}}}_{CR},\ [\hat{\mathscr{H}}_\Delta,\hat{\tilde{\mathscr{H}}}_{CR}]=\Delta\hat{\mathscr{H}}_{CR},\quad (8)$$

where

$$\hat{\tilde{\mathscr{H}}}_{CR}=\sum b_{ij}(\hat{I}_i{}^+\hat{S}_j{}^- - \hat{I}_i{}^-\hat{S}_j{}^+)$$

in the case of pure spin magnetism (see Secs. 1 and 2), and in the more general case it is defined in similar fashion with the aid of the operators $\hat{P}^i_\alpha$. Using (8), we can prove by differentiating with respect to t the following identity:

$$\exp(i\hat{\mathscr{H}}_\Delta t)\hat{\mathscr{H}}_{CR}\exp(-i\hat{\mathscr{H}}_\Delta t)\equiv\hat{\mathscr{H}}_{CR}\cos\Delta t+i\hat{\tilde{\mathscr{H}}}_{CR}\sin\Delta t,$$

and then transform (7) into

$$\langle\hat{\mathscr{H}}_d'+\hat{\mathscr{H}}_{CR}\rangle(t)=-C\beta_d\{\mathrm{Sp}\,\hat{\mathscr{H}}_d'^2+\mathrm{Sp}\,\hat{\mathscr{H}}_{CR}^2\,\mathrm{Re}\,[F(t)\exp i\Delta t]\}, \qquad (9)$$

where

$$F(t)=(\mathrm{Sp}\,\hat{\mathscr{H}}_{CR}^2)^{-1}\,\mathrm{Sp}\,\{\hat{\mathscr{H}}_{CR}\exp(i\hat{\mathscr{H}}_d't)\hat{\mathscr{H}}_{CR}\times\exp(-i\hat{\mathscr{H}}_d't)+\hat{\mathscr{H}}_{CR}\exp(i\hat{\mathscr{H}}_d't)\hat{\hat{\mathscr{H}}}_{CR}\exp(-i\hat{\mathscr{H}}_d't)\}. \qquad (10)$$

Formula (10) is similar in structure to the known expression for the free-induction damping signal (see, e.g., [11]). In the general case $F(t)$ is a complex function of the time ($\mathrm{Re}F(t)$ is even and $\mathrm{Im}F(t)$ is odd), with $F(0)=1$ and $F(\infty)\to 0$. In the course of the evolution, the first term in the right-hand side of (9) remains unchanged, whereas the second term undergoes damped oscillations with frequency and ultimately vanishes.

On the other hand, in view of the conservation of the total energy $\langle\hat{\mathscr{H}}_\Delta+\hat{\mathscr{H}}_d'+\hat{\mathscr{H}}_{CR}\rangle$, the evolution of $\langle\hat{\mathscr{H}}_\Delta\rangle(t)$ takes the form

$$\langle\hat{\mathscr{H}}_\Delta\rangle(t)=-C\beta_d\,\mathrm{Sp}\,\hat{\mathscr{H}}_{CR}^2\{1-\mathrm{Re}[F(t)\exp i\Delta t]\}, \qquad (11)$$

i.e., during the course of the process the difference energy experiences the same oscillations with damping, increasing thereby from zero to the value $\langle\hat{\mathscr{H}}_{CR}\rangle(0)$, which leads in turn to a corresponding change of that part of the longitudinal magnetization of the material which is proportional to $\langle\hat{\mathscr{H}}_\Delta\rangle$. The entire process as a whole can thus be represented as oscillating energy exchange between these subsystems $\hat{\mathscr{H}}_{CR}$ and $\hat{\mathscr{H}}_\Delta$, which terminates by energy transfer from $\hat{\mathscr{H}}_{CR}$ to $\mathscr{H}_\Delta$ and with formation of a quasi-equilibrium "difference" pool. The oscillation damping time can be estimated by expanding $F(t)$ in powers of t, where the traces that enter in the expansion up to t^2 can be directly calculated by a known method, [10] and the dominant lattice sum in the coefficient of t^2 can be separated and estimated in analogy with[7]. Calculation shows that for undiluted spin systems the damping time is of the order of ω_d^{-1}.

We introduce now the "hindrance factor" $\varepsilon=\mathrm{Tr}\hat{\mathscr{H}}_{CR}^2/\mathrm{Sp}\,\hat{\mathscr{H}}_d'^2$, which is connected with the rate of the CR at zero detuning: if $W_{CR}(0)\sim\omega_d$, then ε can reach a value ~ 1, and in the case $W_{CR}(0)\ll\omega_d$ ("forbidden CR") we have $\epsilon\ll 1$. The calculation of ε can be carried out by standard methods (see, e.g., [10]).

It follows from (9)–(11) that the amplitude of the considered transient process turns out to be proportional to $\mathrm{Tr}\,\hat{\mathscr{H}}_{CR}^2$, i.e., it depends directly on the value of ε, whereas the characteristic time of this process does not depend on ε at all and coincides with the time of formation of the spin temperatures.[2)] The latter circumstance is explained by the fact that the mismatch of the oscillation phases corresponding to different pairs of spins is due to the same interaction $\hat{\mathscr{H}}_d'$ which leads to the damping of the free-induction signal.

The Δ pool produced after the fast transient process can be characterized by a temperature β_Δ^{-1}. We note that a similar transient was observed in systems with one sort of spins following sudden application of a magnetic field in the laboratory[7] and rotating[12] coordinate systems, and is attributed in[3,4,8] to unification of the subsystems $\hat{\mathscr{H}}_d''$ and $\hat{\mathscr{H}}_z$. Here we have in fact generalized these results to include the case of CR.

So far we have considered the case $|\Delta|\gg\omega_d$. When this condition is violated, the transient process loses its vibrational character and its description becomes more complicated. Nonetheless, it can be shown (proceeding in analogy with[7]) that in this case, too, the characteristic time of the process is of the order of ω_d^{-1} and does not depend on ε. However, the conclusion that the entire energy $\langle\hat{\mathscr{H}}_{CR}\rangle$ goes over into $\langle\hat{\mathscr{H}}_\Delta\rangle$ is no longer valid here, and it is necessary to take into account the energy exchange between all three subsystems $\hat{\mathscr{H}}_d'$, $\hat{\mathscr{H}}_{CR}$ and $\hat{\mathscr{H}}_\Delta$. A similar situation for one sort of spins was considered in[13].

4. SECOND STATE OF TRANSIENT PROCESS: CROSS RELAXATION PROPER

The next stage in the establishment of the equilibrium is the relatively slow equalization of the temperatures β_Δ^{-1} and β_d^{-1}, i.e., the CR in the proper meaning of the word, and at each instant of time the state of the system can be described by a density matrix in the form

$$\hat{\rho}=C[1-\beta_\Delta(t)\hat{\mathscr{H}}_\Delta-\beta_d(t)\hat{\mathscr{H}}_d']. \qquad (12)$$

We note that in accordance with the idea of unifying the subsystems $\hat{\mathscr{H}}_\Delta$ and $\hat{\mathscr{H}}_{CR}$ into a single energy pool (in analogy with[3,8]), the second term in the square brackets of (12) should be written in the form $\beta_\Delta(t)\,(\hat{\mathscr{H}}_\Delta+\hat{\mathscr{H}}_{CR})$; however, within the framework of the approximation used by us, which is connected with separating the secular part of the DD interaction accurate to first-order perturbation theory, this addition of the term $\hat{\mathscr{H}}_{CR}$ does not influence in any way the result of the subsequent calculation of $W_{CR}(\Delta)$.[3)] We emphasize that if the second order of perturbation theory were to be taken into account, it would be necessary to add to the term $\hat{\mathscr{H}}_\Delta$ in (12) only that part of $\hat{\mathscr{H}}_{CR}$ which causes a small shift of the resonant frequency of the Δ subsystem (cf. [4]).

On the basis of formula (12), we shall use the nonequilibrium statistical operator method[14] to calculate the CR probability $W_{CR}(\Delta)$. In the case $\varepsilon\ll 1$, expression (12) is valid for all Δ (including $\Delta=0$), while in the case $\varepsilon\sim 1$ it describes the quasi-equilibrium only for detunings $|\Delta|\gg\omega_d$.[3] Proceeding in standard fashion we obtain for $W_{CR}(\Delta)$ an expression that can be written, after comparison with (10) in the form

$$W_{CR}(\Delta)=\frac{1}{2}(\mathrm{Sp}\,\hat{N}_\Delta^2)^{-1}\,\mathrm{Sp}\,\hat{\mathscr{H}}_{CR}^2\int_{-\infty}^{\infty}F(t)\exp(i\Delta t)\,dt. \qquad (13)$$

Thus, it turns out that the function $F(t)$, which characterizes the damping of the oscillations after a sudden introduction of the detuning, and the shape of the "CR line" $W_{CR}(\Delta)$ are related by a Fourier transformation and contain the same information. This very significant conclusion shows that from the form of the transient process observed at one fixed value of Δ it is possible to establish the $W_{CR}(\Delta)$ dependence in the entire range of detunings, just as the signal of the free-induction damping makes it possible to determine the magnetic-resonance line shape. When drawing this conclusion, we must stipulate that it is valid under two significant restrictions. First, we are dealing always with a function $F(t)$ corresponding to the turning-on of sufficiently large detunings $|\Delta| \gg \omega_d$; in the opposite case, as mentioned in Sec. 3, an additional coupling arises between $\langle \hat{\mathscr{H}}_{\mathrm{CR}} \rangle$ and $\langle \hat{\mathscr{H}}'_d \rangle$, and distorts the form of the fast transient. Second, at $\varepsilon \sim 1$ we are not justified in using the spin-temperature concept in the derivation of (13) in the region $|\Delta| \lesssim \omega_d$, so that the use of the Fourier transform of the function $F(t)$ is valid in this case only for the calculation of sufficiently remote wings of the "line" $W_{\mathrm{CR}}(\Delta)$. This was precisely the situation with the analysis of the fast transient accompanying the turning on of the field H_0 in systems with spins of one sort. [3] To the contrary, at $\epsilon \ll 1$ it is possible to determine from the form of $F(t)$ the entire "CR line," including the region of small detunings.

We note that the presence of $\mathrm{Im}F(t)$ leads to the appearance of a certain asymmetry of the function $W_{\mathrm{CR}}(\Delta)$, in contrast to the case of pure spin magnetism.

The inhomogeneous broadening of the magnetic-resonance lines leads to an accelerated damping of the oscillations in (9) and (11), on account of interference between the spin packets with different values of Δ. [4)] Nonetheless, if we add to $\hat{\mathscr{H}}'_d$ secular parts of the interactions that cause the inhomogeneity, then formulas (9)–(11) remain in force. However, the quasi-equilibrium that corresponds to the density matrix (12) does not set in immediately after that: it requires the completion of the spectral diffusion within the lines (see, e.g., [5]). If this process is faster than the CR of interest to us (this is always the case if Δ is sufficiently large), then formula (13) also remains in force. If furthermore the inhomogeneous broadening of the initial lines predominates, it is precisely this broadening which determines the shapes of the plots of $F(t)$ and $W_{\mathrm{CR}}(\Delta)$.

5. SCHEME OF A POSSIBLE EXPERIMENT

An experiment aimed at observing the described transient processes can be imagined in the following manner.

A. A sample containing nuclear or electron spins with a non-equidistant magnetic-resonance spectrum is placed in a field $\mathbf{H}_0$, the magnitude and direction of which ensure equality of the frequencies of any two transitions of the spectrum, $\omega_I = \omega_S = \omega_0 \gg \omega_d$.

B. After thermal equilibrium is reached in the field H_0, the transitions I and S are subjected simultaneously to adiabatic demagnetization in a rotating coordinate frame. As a result, the Zeeman temperatures of these transitions, β_I^{-1} and β_S^{-1}, become infinite, whereas the reciprocal temperature of the DDP reaches a value $\beta_d(0) \sim \beta_0 \omega_0/\omega_d$, where β_0^{-1} is the lattice temperature. The adiabatic demagnetization in a rotating coordinate system is not needed here in principle, and is carried out only to increase the average energy stored in the term $\hat{\mathscr{H}}_{\mathrm{CR}}$.

C. At the instant $t = 0$, the magnitude or orientation of $\mathbf{H}_0$ are changed rapidly in such a way that a detuning $|\Delta| \gg \omega_d$ is produced between the frequencies ω_I and ω_S. The jump should be carried out within a time shorter than Δ^{-1}.

D. Immediately after the onset of the detuning, an oscillatory energy transfer begins between $\hat{\mathscr{H}}_{\mathrm{CR}}$ and $\hat{\mathscr{H}}_\Delta$, described by formulas (9)–(11). We consider the quantities $\langle \hat{N}_I \rangle$ and $\langle \hat{N}_S \rangle$, which we shall call the polarizations of the transitions of I and S, in analogy with $\langle \hat{I}_z \rangle$ and $\langle \hat{S}_z \rangle$ in the pure spin case. (We emphasize that the term "population difference" cannot be used here, since quasi-equilibrium has not yet been reached.) It is obvious that $\langle \hat{N}_I \rangle - \langle \hat{N}_S \rangle = k\langle \hat{\mathscr{H}}_\Delta \rangle/\Delta$, where $k = 2$ or 3, depending on the presence of a common level for the I and S transitions (see Sec. 2). At the same time, $\langle \hat{N}_I \rangle + \langle \hat{N}_S \rangle = \mathrm{const} = 0$ (the last equality is the consequence of the adiabatic demagnetization in the rotating coordinate system), so that $\langle \hat{N}_I \rangle = -\langle \hat{N}_S \rangle = k\langle \hat{\mathscr{H}}_\Delta \rangle/2\Delta$. It is clear therefore that to register the change $\langle \hat{\mathscr{H}}_\Delta \rangle$ of interest to us it suffices to follow the polarization of one of the transitions, I or S (for example, by measuring it by resonant methods at the frequency ω_I or ω_S).

E. After the damping of the oscillations, as follows from (11) and (12), the value $\beta_\Delta = \beta_d(0)\,\mathrm{Sp}\,\hat{\mathscr{H}}^2_{\mathrm{CR}}/\mathrm{Tr}\,\hat{\mathscr{H}}^2_\Delta \sim \beta_d(0)\,\varepsilon\,\omega_d^2/\Delta^2$ is established. Taking into account the estimate for $\beta_d(0)$ it follows therefore that $\beta_I = -\beta_S \sim \pm\beta_0\varepsilon\omega_d/\Delta$. Thus, at $\varepsilon \sim 1$ and $|\Delta| \sim (5\text{–}10)\omega_d$, the resultant polarization of the transitions I and S constitutes an appreciable fraction of its equilibrium value and can be easily measured. In the case $\varepsilon \ll 1$ ("forbidden" CR), the detection of the effect is made very difficult (although still not excluded).

F. Finally, the last stage of the experiment can be the registration of the relatively slow process of the "CR proper"; this part of the experiment provides in principle no new information and can be regarded as a control experiment.

The entire experiment should be carried out within a time much shorter than the spin-lattice relaxation time.

The described procedure is not the only one possible. Alternative variants may include, for example, direct observation of the longitudinal magnetization after a fast variation of Δ, or else the determination of the stationary longitudinal-absorption spectrum at low frequencies. In the latter case, as shown in [15], a characteristic maximum should be observed at the frequency Δ, with a shape that reflects the form of the function $W_{\mathrm{CR}}(\Delta)$; it is clear that this "line" is due to the fast relaxation process considered by us and is related to $F(t)$ by a Fourier transformation. [5)]

Thus, observation of the described effects is perfectly realistic. In addition to its fundamental significance, such experiments yield a new method of studying spin-spin interactions (and, in particular, the CR "line" shape)—a method in the spirt of modern pulsed Fourier spectroscopy.

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[1)]For simplicity we shall refer only to DD interactions, and accordingly to the dipole–dipole pool (DDP), although all the results can be easily generalized also to other types of spin–spin interactions, such as exchange interactions.

[2)]We note that this conclusion contradicts the idea of treating the subsystem $\hat{\mathcal{H}}_{CR}$ as a separate quasi-equilibrium pool.[6]

[3)]This circumstance was pointed out to us by L. L. Buishvili.

[4)]This damping, of course, is not irreversible, but here, at least in principle, experiments of a spin-echo type are possible.

[5)]At the same time, this "line" can be regarded as a result of induced two-spin transitions: in this sense it is the analog of the low-frequency "Van Vleck satellites" in magnetic-resonance spectra.

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Stratification of a heated electron-hole plasma

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It is found that the homogeneous density distribution of a quasineutral electron-hole plasma becomes unstable when the plasma is uniformly heated by photogeneration or in an electric field, even if the condition for ordinary superheat instability is not fulfilled and external fluxes (temperature and concentration gradients, or a current) are absent. The instability is of an aperiodic nature and arises with respect to perturbations with a characteristic wave vector $k_0=(Ll)^{-1/2}$, where L is the bipolar diffusion length and l is the hot electron cooling length. It is shown in the "hydrodynamic" approximation that at $L>l$ stratification of the plasma occurs when the hot-electron momentum is scattered by charged centers, irrespective of the mechanism of the electron energy relaxation. From an analysis of the nonlinear one-dimensional problem it follows that the stable distributions of the carrier density and of the carrier temperature are those having the form of large-amplitude oscillations with a period of the order of $(Ll)^{1/2}$. The stratification mechanism considered may be one of the causes of the "drop formation" observed experimentally in a nonequilibrium electron-hole plasma of a semiconductor.

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1. INTRODUCTION. STRATIFICATION MECHANISM

In homogeneous systems that are disturbed by external actions out of their thermodynamic equilibrium state, ordered inhomogeneous stationary states are frequently produced.[1] Thus, for example, superheat instability, which leads to a non-single-valued current-voltage characteristic, produces in semiconductors an inhomogeneous distribution of the electric field or of the current density.[2,3] The formation of such inhomogeneous states can be connected with the presence of not only external but also "latent" (with respect to one of the parameters) negative differential resistance.[4,5] When external conditions produce temperature gradients or carrier-density gradients, the onset or the ordered structure takes place under less stringent requirements with respect to the semiconductor parameters.[6,7] In particular, in the presence of a temperature gradient, static spatially-periodic distributions are produced both in liquids (the Benard phenomenon) and in systems of hot electrons in semiconductors.[7] In this paper we consider an instability homogeneously heated by a quasi-neutral electron-hole plasma or a weakly-ionized gas, leading to stratification even in those cases when the conditions of the superheat instability are not satisfied and there are no fluxes (gradients of the temperature and of the carrier density) as specified by the external conditions.

The gist of the considered stratification mechanism of an electron-hole plasma (weakly ionized gas) consists in the following. Assume that electron-hole pairs are homogeneously generated in a semiconductor by photo-