Magnetic resonance in a solid in the case of a narrow asymmetric spin-lattice interaction line

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Magnetic resonance in a solid is considered for an arbitrary shape and width of the spin-phonon interaction line $W(\omega)$. A system of equations for the evolution of the Zeeman and non-Zeeman temperatures β_z^{-1} and β_{nz}^{-1} is obtained from simple physical considerations and is solved in the case of spin-lattice relaxation and for magnetoresonance saturation. It is shown that the physical picture of both processes differs substantially from the traditional one if the $W(\omega)$ line is narrow compared with the magnetoresonance line $P_0(\omega)$ and (or) is asymmetric with respect to its center ω_0. In particular, in the stationary saturation regime it is possible to have not only a strong cooling of the non-Zeeman (low-frequency) reservoir ($|\beta_{nz}|\gg\beta_L$, where β_L^{-1} is the lattice temperature), but also a noticeable "superheating" of the Zeeman (high-frequency) reservoir ($\beta_z<0$, $|\beta_z|\sim\beta_L$), a fact manifest, e.g., in a considerable inversion of the basic (central) part of the magnetoresonance line $P(\omega)$. Estimates demonstrate that it is realistic to expect experimental observation of the predicted effects and to use them, in particular, to investigate interaction in solid-state spin systems.

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1. INTRODUCTION

It is usually assumed[1-3] that in a solid-state spin system its two constituent quasi-equilibrium subsystems, the Zeeman (high-frequency) and the non-Zeeman (low-frequency) subsystems, with Hamiltonians $\hat{\mathscr{H}}_z$ and $\hat{\mathscr{H}}_{nz}$, relax into the lattice independently of each other, each following a single exponential curve. This, however, is not always the case. Let the spin-lattice relaxation be effected via a direct interaction of the spins with the phonons in the vicinity of the Zeeman frequency ω_0, as is the case, e.g., in electron paramagnetic resonance (EPR), inasmuch as the $\hat{\mathscr{H}}_{nz}$ reservoir does not have its own separate contact with the lattice; we shall consider hereafter precisely this case and designate the spin-phonon interaction line by $W(\omega)$. Then the independent singly exponential spin-lattice relaxation of each of the subsystems $\hat{\mathscr{H}}_z$ and $\hat{\mathscr{H}}_{nz}$ follows from the symmetry of the line $W(\omega)$ about ω_0. This statement follows, in particular, from the paper of Buishvili and Zviadadze,[4] who considered magnetic resonance broadened by dipole-dipole interactions ($\hat{\mathscr{H}}_{nz}\equiv\hat{\mathscr{H}}_d$). Since the spin-phonon line is slightly asymmetrical about ω_0 for direct (single-phonon) relaxation processes ($W(\omega)\propto\omega^2$), a second exponential appears in the case of spin-lattice relaxation of each of the subsystems, and in the regime of stationary saturation at the frequency ω_0, the reservoir $\hat{\mathscr{H}}_d$ is somewhat cooled (its temperature β_d^{-1} decreases to one-third the lattice temperature β_L^{-1}), something that should not happen in accordance with the accepted theory of magnetic-resonance saturation in solids.[1,2] These effects, however, are small and were disregarded in most of the succeeding investigations and reviews (Refs. 3, 5, 6).[1)]

There exist nevertheless substances in which the assumed picture of the spin-lattice relaxation may become quite greatly distorted. First among them are spin systems in which the magnetic-resonance line is broadened not by the dipole-dipole interaction, and moreover, by no spin-spin interactions at all, but is formed of an aggregate of narrow lines close in frequency, due to the so-called "spin packets" (this is typical of EPR). If the packets are subjected to effective cross relaxation (i.e., the latter predominates over their relaxation to the lattice), the aggregate inhomogeneous line behaves in magnetic resonance like a homogeneous line, i.e., it can be described by the two temperatures β_z^{-1} and β_{nz}^{-1} of the reservoirs $\hat{\mathscr{H}}_z$ and $\hat{\mathscr{H}}_{nz}$. The mean frequency ω_{nz} of the reservoir $\hat{\mathscr{H}}_{nz}$ produced by the cross relaxation and responsible for the broadening of the magnetic resonance is equal to the square root of the second moment of the frequencies of the packets relative to the center of gravity ω_0.[3,6,8-10] It is important that the spin-lattice times W_i^{-1} of different packets can differ greatly from one another—both because of the difference in their spin-phonon parameters and because of the absence of rapidly relaxing resonant admixtures. This means in turn that the spin-phonon line $W(\omega)$ may turn out to be quite narrow compared with the magnetic-resonance line $P_0(\omega)$ and (or) strongly asymmetrical with respect to its center ω_0.

This paper deals with physical manifestations of such a situation. We describe the behavior of a spin system in spin-lattice relaxation and in magnetic-resonant saturation. It is shown that new experimentally observable effects appear in both processes. In particular, in the regime of stationary saturation it is possible to obtain not only deep cooling of the low-frequency $\hat{\mathscr{H}}$ reservoir $\hat{\mathscr{H}}_{nz}$ ($|\beta_{nz}|\gg\beta_L$), but also noticeable "superheating" (inversion) of the high frequency reservoir ($\beta_z<0$, $|\beta_z|\sim\beta_L$); this manifests itself, e.g., in a considerable inversion of the fundamental (central) part of the magnetic-resonance line (a phenomenon forbidden in the accepted theory[1-3]). An experimental study of these effects, besides their possible direct use, can yield extensive information on the character of the spin-spin and spin-lattice interactions in solids.

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2. SPIN LATTICE RELAXATION

In the description of the relaxation process due to the interaction of spins with phonons at the frequency $\omega = \omega_0 + \delta$ ($\delta \ll \omega$), an analogy should take place with the saturation of the magnetic resonance at the frequency ω, inasmuch as in both cases an interaction takes place with a thermostat (phonons or photons) of given temperature (β_L^{-1} and the infinite radiation temperature, respectively). In both cases the process reduces to a tendency of the spin temperature $[\beta(\omega)]^{-1}$ at the frequency ω (Refs. 6, 7, 10) to approach the thermostat temperature; here

$$\beta(\omega) = \omega^{-1}(\omega_0\beta_z + \delta\beta_{nz}).$$

These arguments lead to a pair of equations for the spin-lattice relaxation at each frequency ω:

$$\dot{\beta}_z = \frac{\omega_{nz}^2}{\delta\omega_0}\dot{\beta}_{nz} = -\frac{\omega}{\omega_0}W(\omega)[\beta(\omega) - \beta_L],$$

and integration with respect to ω yields

$$\begin{aligned} \dot{\beta}_z &= -W(\beta_z - \beta_L) - \frac{\Delta_1}{\omega_0}W(\beta_{nz} - \beta_L), \\ \dot{\beta}_{nz} &= -\frac{\Delta_1\omega_0}{\omega_{nz}^2}W(\beta_z - \beta_L) - \frac{\Delta_2^2}{\omega_{nz}^2}W(\beta_{nz} - \beta_L), \end{aligned} \qquad (1)$$

where

$$W = \int W(\omega)\,d\omega, \quad \Delta_1 = \frac{1}{W}\int \delta W(\omega)\,d\omega,$$

$$\Delta_2^2 = \frac{1}{W}\int \delta^2 W(\omega)\,d\omega, \quad \delta = \omega - \omega_0, \quad \Delta_1^2 \leqslant \Delta_2^2, \quad \Delta_1\Delta_2 \geqslant 0.$$

The system (1) corresponds to the equations of the paper of Buishvili and Zviadadze,[4] which were rigorously derived for dipole-dipole broadening of the magnetic resonance. The solution of Eqs. (1) is of the form

$$\begin{aligned} \beta_z &= Z'\exp(r_1 t) + Z''\exp(r_2 t) + \beta_L, \\ \beta_{nz} &= D'\exp(r_1 t) + D''\exp(r_2 t) + \beta_L, \end{aligned} \qquad (2)$$

where $r_1 < 0$ and $r_2 < 0$. In the usual case $\Delta_1 = 0$ [e.q., the $W(\omega)$ line is symmetrical about ω_0], the reciprocal temperatures β_z and β_{nz} tend to the value β_L independently, along a single exponential, with respective rates $W_z = -r_1 = W$, $W_{nz} = -r_2 = W\Delta_2^2/\omega_{nz}^2$, $Z'' = D' = 0$. As a rule, the line $W(\omega)$ is broad, i.e., $\Delta_2^2 \geqslant \omega_{nz}^2$, meaning $W_{nz} \geqslant W_z$ (thus, for dipole-dipole broadening of magnetic resonance we have $\Delta_2^2 = 3\omega_{nz}^2$, Refs. 4–6). When Δ_1 deviates from zero and approaches Δ_2, a second "crossover" exponential appears in the evolution of the temperature, so that each of the quantities β_z and β_{nz} becomes already dependent on both their initial values. The rates $|r_1|$ and $|r_2|$ move away from each other and tend to the values $r_1 = 0$ and $r_2 = -W(1 + \Delta_2^2/\omega_{nz}^2)$ corresponding to the limiting case $\Delta_1 = \Delta_2$—an infinitely narrow (monochromatic) line $W(\omega)$, when a definite connection between them, given by an exponential with argument $r_2 t$, common to both temperatures, is established between them, and the lattice temperature β_L^{-1} is reached only after an infinite time. This is precisely the case which is fully analogous to saturation of magnetic resonance with a detuning $\Delta = \Delta_1 = \Delta_2$. A new picture of the spin-lattice relaxation can be expected for a line $W(\omega)$ that is sufficiently narrow and (or) asymmetrical with respect to ω_0.

We introduce the line-narrowness parameter

$$\alpha = 4\frac{(\Delta_2^2 - \Delta_1^2)\omega_{nz}^2}{(\Delta_2^2 + \omega_{nz}^2)^2}.$$

Under the condition $\alpha \ll 1$ it follows from (1) and (2) that

$$r_1 \approx -kW, \quad r_2 \approx -W(1 + \Delta_2^2/\omega_{nz}^2 - k), \qquad (3)$$

where

$$k = (\Delta_2^2 - \Delta_1^2)/(\Delta_2^2 + \omega_{nz}^2), \quad 0 \leqslant k \leqslant 1.$$

The amplitudes of the exponentials in (2) are given by

$$\begin{aligned} Z' &\approx \frac{\Delta_1}{\omega_0}\frac{1}{k-1}D' \\ &\approx \frac{\Delta_1^2}{\Delta_1^2 + \omega_{nz}^2(1-k)^2}\left\{[\beta_z(0) - \beta_L] - \frac{\omega_{nz}^2(1-k)}{\Delta_1\omega_0}[\beta_{nz}(0) - \beta_L]\right\}, \end{aligned} \qquad (4)$$

$$\begin{aligned} Z'' &\approx \frac{\omega_{nz}^2}{\Delta_1\omega_0}(1-k)D'' \\ &\approx \frac{\omega_{nz}^2(1-k)^2}{\Delta_1^2 + \omega_{nz}^2(1-k)^2}\left\{[\beta_z(0) - \beta_L] + \frac{\Delta_1}{\omega_0(1-k)}[\beta_{nz}(0) - \beta_L]\right\}, \end{aligned}$$

where $\beta_z(0)$ and $\beta_{nz}(0)$ pertain to the initial instant of the relaxation process. It follows from (3) that $|r_1| \ll |r_2|$, so that we can separate the first stage of the relaxation, which is completed after a time $\theta \sim |r_2|^{-1}$ and which lasts long enough; we then have from (2)

$$\beta_z(\theta) \approx Z' + \beta_L, \quad \beta_{nz}(\theta) \approx D' + \beta_L.$$

Let the initial state $\beta_z(0)$ and $\beta_{nz}(0)$ be produced by saturation of the equilibrium system, and let the saturating pulse be short enough for the saturation to take place practically isolated from the lattice. We shall dwell on two cases. Assume that in the first case the pulse had a detuning Δ ($|\Delta|\omega_0 \gg \omega_{nz}^2$), so that[1-3]

$$\beta_z(0) - \beta_L \approx -\frac{\omega_{nz}^2}{\Delta^2 + \omega_{nz}^2}\beta_L, \quad \beta_{nz}(0) - \beta_L \approx \beta_{nz}(0) \approx -\frac{\Delta\omega_0}{\Delta^2 + \omega_{nz}^2}\beta_L. \qquad (5)$$

When the pulse is then turned off, the signal $P(\omega_0, t)$, which is proportional to $\beta_z(t)$, at the center of the magnetic resonance, can continue to decrease during the first stage of the relaxation, despite its usual monotonic growth when the equilibrium is established, as is seen from (4′) and (5), inasmuch as under the condition $\Delta\Delta_1 < (k-1)\omega_{nz}^2 < 0$ we have $Z'' > 0$. We note that under this condition, in the limit of an infinitely narrow line $W(\omega)$ ($k \to 0$) its peak is located in that frequency region where as a result of saturation the signal $P(\omega)$ has exceeded the equilibrium value.[2,3,11] To estimate the decrease of $P(\omega_0, t)$ following the relaxation, we construct a line consisting of five equidistant infinitely narrow magnetic-resonance lines of equal height (corresponding to five "spin packets"), subjected to effective cross relaxation, with the spin-lattice relaxation proceeding faster (say, by 10 times) in one of the other packets than in the four remaining ones; it is this model which we shall use hereafter. For such a line calculation yields $\omega_{nz} = 2^{1/2}a$ (a is the distance between the centers of the packets), $|\Delta_1| \approx 1.3\,a$, $\Delta_2 = 1.8\,a$, $k \approx 0.31$, $r_2/r_1 \approx 8$, and after the cessation of the saturation of the outermost packet, which is symmetrical to the one that relaxes rapidly, the first stage proceeds in such a way that the signal from the middle (central) packet continues to decrease by another 10% from its equilibrium value; this is patently sufficient to be observable.

In the second case we consider the spin-lattice relaxation after saturation of the line center ω_0. The saturation yields $\beta_z(0)\approx 0$, $\beta_{nz}(0)=\beta_L$, and the subsequent relaxation during the first stage leads, in accord with (4), to the state

$$\beta_{nz}(\theta)\approx D'+\beta_L\approx\left[1+\frac{\Delta_1\omega_0(1-k)}{\Delta_1^2+\omega_{nz}^2(1-k)^2}\right]\beta_L, \tag{6}$$

i.e., at $\Delta_1\neq 0$ and small k we have $|\beta_{nz}(\theta)|\gg\beta_L$. The physical meaning of this effect is clear, for in the limit of an infinitely narrow line $W(\omega)$ it is analogous to the results of the first stage of the saturation of the equilibrium line of magnetic resonance with detuning $\Delta=-\omega_{nz}^2/\Delta_1$, and the line $P(\omega,\theta)$ corresponding to expression (6) is analogous to the magnetic-resonance line in saturation,[3,11] including the inversion section on the wing opposite to Δ_1, and the section where the equilibrium line is higher on the other wing. For our model of five lines, calculation with the aid of (6) yields

$$|\beta_{nz}(\theta)|/\beta_L\approx 0.48\omega_0/\omega_{nz}.$$

Thus, this effect is also large enough to be observable.

3. MAGNETIC-RESONANT SATURATION

We turn now to manifestations of spin-lattice relaxation with line $W(\omega)$ of arbitrary width and shape, under the action of a saturating field with a detuning Δ. The equations for this simultaneous interaction of a spin system with two thermostats having different temperatures can be easily written if one knows the equation for each process separately:

$$\begin{gathered}\dot\beta_z=-W(\beta_z-\beta_L)-\frac{\Delta_1}{\omega_0}W(\beta_{nz}-\beta_L)-p\left(\beta_z+\frac{\Delta}{\omega_0}\beta_{nz}\right),\\ \dot\beta_{nz}=-\frac{\Delta_1\omega_0}{\omega_{nz}^2}W(\beta_z-\beta_L)-\frac{\Delta_2^2}{\omega_{nz}^2}W(\beta_{nz}-\beta_L)-\frac{\Delta\omega_0}{\omega_{nz}^2}p\left(\beta_z+\frac{\Delta}{\omega_0}\beta_{nz}\right),\end{gathered} \tag{7}$$

where p is the probability of the interaction with the saturating field per unit time. Let, as is usually assumed, the saturation be sufficiently strong, i.e., $p\gg W$. Then integration of the system (7) yields for the arguments r_1t and r_2t of the exponential approaches to the stationary state,

$$r_1\approx-p\left(1+\frac{\Delta^2}{\omega_{nz}^2}\right),\quad r_2\approx-W\frac{(\Delta-\Delta_1)^2+\Delta_2^2-\Delta_1^2}{\Delta^2+\omega_{nz}^2}, \tag{8}$$

and it is clear that $|r_1|\gg|r_2|$. As for the stationary values $\beta_z(\infty)$, $\beta_{nz}(\infty)$, we have, putting $\dot\beta_z=\dot\beta_{nz}=0$ in (7),

$$\frac{\beta_z(\infty)}{\beta_L}\approx\frac{\Delta(\Delta-\Delta_1)}{(\Delta-\Delta_1)^2+\Delta_2^2-\Delta_1^2}, \tag{9}$$

$$\frac{\beta_{nz}(\infty)}{\beta_L}\approx\frac{(\Delta_1-\Delta)\omega_0+\Delta_2^2-\Delta_1^2}{(\Delta-\Delta_1)^2+\Delta_2^2-\Delta_1^2}\approx\frac{(\Delta_1-\Delta)\omega_0}{(\Delta-\Delta_1)^2+\Delta_2^2-\Delta_1^2}. \tag{9'}$$

We note that (9′) can be regarded as the analog of the result of stationary saturation of a magnetic resonance line in which the center is located at a frequency $\omega_0+\Delta_1$ and

$$\omega_{nz}^2W_{nz}/W_z=\Delta_2^2-\Delta_1^2.$$

We consider now the manifestations of the stationary state in various cases.

1. Let the line $W(\omega)$ have a zero mean value, $\Delta_1=0$ (e.g., let it be symmetrical with respect to ω_0), but relatively narrow: $\Delta_2^2<\omega_{nz}^2$. Then, comparing (9′) with (5), we note that in contrast to the usual picture the effects of the abrupt increase of $|\beta_{nz}|$ and of the characteristic asymmetry of the magnetic-resonance line, which appear when the line is saturated with a detuning Δ,[3,11] turn out to be stronger not during the first stage but during the second, after the stationary state is established (after the so-called "spin-lattice relaxation in the rotating coordinate system"[2]). This corresponds to an unusual ratio of the spin-lattice rates of the two reservoirs: $W_{nz}/W_z=\Delta_2^2/\omega_{nz}^2<1$, as seen from (1), at $\Delta_1=0$.

2. Let now $\Delta_1\neq 0$, and let the saturating field be applied at the center of the magnetic resonance, $\Delta=0$. It follows then from (9′) that $\beta_{nz}(\infty)/\beta_L\approx\Delta_1\omega_0/\Delta_2^2$, i.e., at $\Delta_2/\Delta_1\ll|\omega_0/\Delta_2|$ the reservoir $\hat{\mathscr{H}}_{nz}$ will be strongly cooled, something that does not happen in accordance with the assumed theory in the case of saturation at the center.[1-3] The effect can be quite large, much larger than that predicted in[4] for dipole-dipole broadened lines. Thus, for the five-line model described above we have $\beta_{nz}(\infty)/\beta_L\approx 0.71\omega_0/\Delta_2$. The magnetic-resonance line $P(\delta)$ becomes sharply antisymmetrical upon stationary saturation: $P(\delta)=(\Delta_1\delta/\Delta_2^2)P_0(\delta)$, where $P_0(\delta)$ is the equilibrium line.

3. We consider now the general case. Trying to reach a maximum of $|\beta_{nz}(\infty)|$, we choose the optimal saturation detuning $\Delta=\tilde\Delta$ such that $(\tilde\Delta-\Delta_1)^2=\Delta_2^2-\Delta_1^2$. Then

$$\tilde\beta_z(\infty)/\beta_L\approx{}^1/_2\tilde\Delta/(\tilde\Delta-\Delta_1), \tag{10}$$

$$\tilde\beta_{nz}(\infty)/\beta_L\approx{}^1/_2\omega_0/(\Delta_1-\tilde\Delta), \tag{10'}$$

i.e.,

$$|\beta_{nz}(\infty)|^{max}/\beta_L\approx{}^1/_2\omega_0/(\Delta_2^2-\Delta_1^2)^{1/2}\gg 1.$$

Thus, the narrower the line $W(\omega)$, the deeper can we cool the reservoir $\hat{\mathscr{H}}_{nz}$ by stationary saturation with optimal detuning. This is natural, inasmuch as for a narrow spin-lattice line $W(\omega)$ and for a saturation frequency close to it the straight line representing the distribution of the reciprocal spin temperatures $\beta(\omega)$ becomes quite steep. Then, however, according to (8) it is necessary to wait all the longer for the establishment of this stationary state. The shift of $|\tilde\beta_{nz}(\infty)|$ as given by (10′) seems exceedingly large; it is unusual,

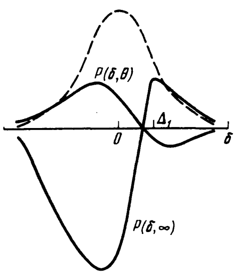

FIG. 1. Magnetic resonance signals in the case of saturation with detuning Δ: $P(\delta,\theta)$—after the first stage (isolated from the lattice), $P(\delta,\infty)$—stationary signal. Dashed curve—equilibrium signal. It is assumed that $\Delta=\frac{2}{3}\Delta_1=2\cdot 10^{-1/2}\Delta_2=3^{-1/2}\omega_{nz}$.

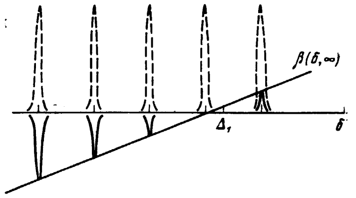

FIG. 2. Magnetic-resonance signal and distribution line of reciprocal spin temperatures $\beta(\delta,\infty)$ in stationary saturation of an aggregate of five lines subjected to effective cross relaxation. Dashed—equilibrium signal. The saturating field is applied to the second line from the right. The rates of the spin-lattice relaxations are respectively $10W$ for the extreme right line and W for the remaining four lines.

however, only if the entire broad magnetic-resonance line is considered. In fact, (10′), as already noted in connection with (9′), can be regarded as the known formula of the theory[1-3,11] for a narrow line $W(\omega)$ separated from the full magnetic-resonance line; the shift of $\hat{\mathscr{H}}_{nz}$, which is determined only by the parameters of the line $W(\omega)$ and is therefore very large, extends gradually over the remaining part of the line, without taking part in the spin-lattice relaxation.

Nonetheless, the stationary increase of $|\beta_{nz}|$ reached according to (10′) is really very large and is limited only by the width of the $W(\omega)$ line. Moreover, (10) leads to the possibility of a phenomenon that is absolutely excluded by the accepted theory—the attainment of a negative Zeeman-reservoir temperature, in the stationary regime, $\beta_z(\infty)<0$. The condition for this, according to (10), is the relation $\Delta\Delta_1>0$, $|\Delta|<|\Delta_1|$, and the magnitude of the effect can be quite large, up to the case $\beta_z(\infty)<-\beta_L$ if the $W(\omega)$ line is narrow enough (when $\Delta_1^2/\Delta_2^2>0.9$). A negative temperature $[\beta_z(\infty)]^{-1}$ means that the basic larger part of the magnetic resonance line, including its center, is inverted before the stationary saturation is reached, and the magnitude of the inversion is appreciable [see Fig. 1 for $\Delta_2=\frac{1}{3}\cdot 10^{1/2}\Delta_1$, $\Delta=\tilde{\Delta}=\frac{2}{3}\Delta_1$, $\beta_z(\infty)=-\beta_L$]. The state of the spin system and the shape of the magnetic-resonance line change abruptly even qualitatively on going from the first saturation state (practically in isolation from the lattice, within the time $\theta\sim|r_1|^{-1}$) to the second stage—the stationary stage: the sign of β_{nz} is reversed and therefore the frequency regions where the line inversion is observed change places (Fig. 1). For the five-line model, the stationary signal $P(\omega)$ and the line representing the distribution of the reciprocal temperatures $\beta(\omega)$ are shown in Fig. 2. Thus, the effect of inversion of the central part of the magnetic-resonance line can be quite large and can lend itself to observation and experiment and to practical use. Observation of the remaining effect is also quite realistic. The objects of the experiments could be solid-state spin systems, in which a noticeable difference between the spin-lattice times is possible within a narrow range of magnetic-resonance frequencies (e.g., radicals in EPR). At sufficiently low temperatures (at which, however, the state of the so-called "photon bottleneck" has not yet been reached[5-7]) the relative efficiency of the spin-spin interactions increases, and this contributes to manifestations of the reservoir $\hat{\mathscr{H}}_{nz}$ and to cross relaxation, i.e., to the onset of the effects described above. Such experiments would make it possible to assess both the degree of efficiency of the cross relaxation and the distribution of the spin-lattice velocities, and consequently also the mechanisms of the spin-phonon interactions.

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[1)] The only exceptions are the studies of the Tbilisi school (see, e.g., Ref. 7).

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