

Dipole-dipole reservoir saturation by nonresonant ultrasound

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Saturation of the dipole-dipole reservoir (DDR) of Cr^{3+} ions in ruby, brought about by direct absorption of the energy of a low-frequency nonresonant ultrasonic field, is observed experimentally for the first time. The dependence of the DDR temperature on the intensity of the longitudinal ultrasonic wave can be described satisfactorily by curve (1). Ultrasonic saturation of the DDR is considered theoretically. The stationary solution of the corresponding kinetic equations is functionally identical to formula (1). A reasonable value of the spin-lattice relaxation time τ_{sl} is obtained by comparing the theory with the experimental results. This indicates the possibility that the ultrasonic DDR saturation technique may be a means of studying spin-lattice processes in paramagnetic crystals.

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Nonresonant methods of investigation of paramagnetic materials have long been known.¹ However, because of their low sensitivity in comparison with resonance methods, they have long failed to achieve widespread use. Lately since it has been possible to increase the sensitivity by one to two orders of magnitude with the aid of the temperature shift of the dipole-dipole reservoir (DDR),² the situation has changed (see, for example, Refs. 3–5). The absorption of a nonresonant alternating magnetic field and the DDR saturation produced by it have been studied in cited works. We have previously⁶ described an experiment on the nonresonant absorption of ultrasound by the spin system of paramagnetic ions Cr^{3+} in ruby under conditions of a DDR temperature shift, at an orientation and a value of the longitudinal magnetic field such that three of the four levels of the chromium ion are equivalent ($\nu_{23} = \nu_{34}$).

The theoretical interpretation of the experimental results was based on the fact that, in the case of identical energy intervals between the spin levels of a paramagnetic particles, a low-frequency sound wave causes direct transitions between the dipole-dipole sublevels into which the states with a given energy of the single-particle spectrum of the system are split. From a comparison of the expression for the second absorption coefficient α , obtained theoretically with account of the considerations set forth above, with the experimentally measured value, a value of the spin-phonon coupling constant was found that is in satisfactory agreement with the data of other methods. This indicates the possibility of obtaining useful information by the method of nonresonant absorption, and is a definite confirmation of the effectiveness of the absorption mechanism considered.

It is clear that if we increase the intensity of the ultrasonic wave, then we can bring about a considerable change in the populations of the dipole-dipole sublevels, i. e., a change in the DDR temperature and, in the final analysis, of the DDR saturation. The experimental and theoretical study of this effect is of interest in itself, and can also give information on the rates of spin-lattice relaxation processes, and would appear to

be a confirmation of the validity of the choice of mechanism for the ultrasound absorption.

2. The experimental investigation of the effect of ultrasonic field on the DDR temperature of Cr^{3+} ions (concentration ~0.05 at. %) under conditions of equidistant energy levels has been carried out in the following manner. At a value of the external magnetic field $H_0 = 1320$ Oe and an angle between the axis of the crystal and the magnetic field $\theta \approx 26^\circ$, the transition frequencies between the levels 2, 3, and 4 of the Cr^{3+} ions become identical ($\nu_{23} = \nu_{34} = 4.7$ GHz; the enumeration of the levels is in the order of increasing energy). Under these conditions, nonresonant absorption of the energy of an alternating magnetic field of low frequency is observed, and from the magnitude of the absorption we can assess the DDR temperature.^{2,4} As an absorption detector, we used an ordinary autodyne. The DDR was saturated by a longitudinal ultrasonic wave propagating along the C_3 axis of the crystal. The measurements were carried out with the use of ultrasound of frequency $\Omega/2\pi = 19619.8$ kHz, which corresponds to mechanical resonance in the sample (the equivalent resonant acoustic resistance $R_a \approx 1.1$ k Ω , $Q \approx 5300$).

To increase the sensitivity, the DDR temperature was first lowered to 0.21 K by non-strictly-resonant microwave pumping of the EPR line Cr^{3+} -ions, corresponding to the transition 2–4, on the low-frequency wing with detuning $\delta/2\pi$ equal to 110 MHz relative to the center of the line. The coefficient of absorption of the ultrasonic field energy by the dipole-dipole reservoir under these conditions amounted to $\alpha \approx 10^{-7}$ cm⁻¹, as reported earlier.⁶ The construction of the measurement cell was described in the same article. The sample was placed in liquid helium at a temperature of $T = 4.2$ K. The temperature of the sample in the presence of the ultrasonic field of maximum intensity¹⁾ (20 W/cm²) did not differ from the temperature of the helium bath, as established by measurement of the time T_1 of the spin-lattice relaxation of the Cr^{3+} ions in the transition ($+\frac{1}{2} \leftrightarrow -\frac{1}{2}$) at $\theta = 0^\circ$ in the presence and absence of the ultrasonic field.²⁾ Within the limits of the measurement accuracy (~5%) there was no difference noted in the values of T_1 (we recall that the temperature de-

pendence of the rate of spin-lattice relaxation is linear in the range 1.4–20 K⁷).

Figure 1 shows the dependence of the ratio of the DDR temperatures T_{dd}^0/T_{dd} (T_{dd}^0 is the DDR temperature in the absence of the ultrasonic field and T_{dd} is its value at ultrasonic saturation), defined as the ratio $A(I)/A(0)$ of the corresponding amplitudes of the non-resonant absorption of the alternating magnetic field energy, on the intensity of the ultrasonic wave in the sample, which was estimated from the measured voltage at the acoustic resonator and its equivalent resonance resistance and Q factors. It is seen that the dependence is satisfactorily described by the curve

$$T_{dd}^0/T_{dd} = (1 + I/I_0)^{-1}, \quad (1)$$

where I is the intensity of the ultrasonic wave in the sample, and $I_0 \approx 2W/cm^2$. In order to eliminate the saturating effect of the alternating magnetic field on the DDR, the quantity $A(I)/A(0)$ was measured by us for alternating magnetic fields of different amplitude h . No dependence of $A(I)/A(0)$ on h was observed, so that we can regard the field that records the DDR temperature to be unsaturated.

It should be pointed out that in the case in which the ultrasonic field occupies only part of the volume of the sample, the ratio T_{dd}^0/T_{dd} should not be less than the value $1 - V_{us}/V$, as is clear from simple considerations. Here V_{us} and V are respectively the volume irradiated by the ultrasonic field and the total volume of the sample. The fact that the experimental data are satisfactorily described by the formula given above indicates that the boundary effects leading to a difference between V_{us} and V are small.

In order to confirm the effectiveness of the previously assumed,⁶ mechanism of direct absorption of the energy of the ultrasonic field by the dipole-dipole reservoir, we carried out the following experiment. At a value of the angle $\theta = 26^\circ$, the magnetic field was changed by jumpwise at a frequency 17 Hz from a value H_{cq} corresponding to equidistant energy levels 2, 3, and 4 of the Cr³⁺ ion, to a value exceeding H_{cq} by 60 Oe. The value of the signal of the nonresonant absorption of energy of the alternating magnetic field of frequency 8 MHz (and consequently of the quantity T_{dd}) was obtained by measuring the 17-Hz amplitude, the signal component at the output of the autodyne. An ultrasonic pulse

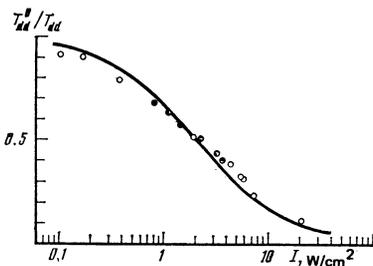


FIG. 1. Dependence of T_{dd}^0/T_{dd} on the intensity of the ultrasonic wave in the sample (T_{dd}^0 is the DDR temperature in the absence of the ultrasonic field and T_{dd} is its value in the case of ultrasonic saturation).

with duration ~ 20 msec was switched on either at the instant when the levels 2, 3, and 4 were equidistant, or at the instant when the levels were not equidistant. In the first case, we observed a strong increase in the DDR temperature, in the second, a much weaker one. This agrees with the considerations set forth previously⁶ and in the first section of this paper.

3. The absorption of the ultrasonic energy by the reservoir of dipole-dipole interactions, which are secular relative to the group of equidistant levels, was previously considered⁶ in the framework of the linear response. The ultrasonic field was assumed to be sufficiently weak that it produced no significant change in the thermodynamic properties of the DDR, the temperature of which was determined by not strictly resonant pumping and therefore this approach was correct. Since our purpose here is to take into consideration the effect of DDR saturation by a sufficiently strong ultrasound, we turn to the method of kinetic equations, which is more suitable for this purpose.

A system of paramagnetic particles possessing a spectrum of n levels, $2S + 1$ of which are equidistant, can be described by the Hamiltonian. $\mathcal{H}_i = \mathcal{H}_0 + \mathcal{H}_i^1$, where

$$\mathcal{H}_0 = \mathcal{H}_s + \mathcal{H}_a + \mathcal{H}_L, \quad \mathcal{H}_i^1 = \mathcal{H}_{sL} + \mathcal{H}_p^i + \mathcal{H}_n^i. \quad (2)$$

The Hamiltonian

$$\mathcal{H}_s = \sum_{j,m=-S}^S \hbar \Delta_s m P_{mm}^j = \hbar \Delta_s S_z \quad (3)$$

describes the equidistant (with a distance $\hbar \Delta_0$ between neighboring energy levels) part of the spectrum of the paramagnetic particle, while the operators are determined by the matrix elements

$$\langle \mu | P_{mm'} | \mu' \rangle = \delta_{\mu\mu'} \delta_{m'm}.$$

on functions that diagonalize the single-particle spectrum.³⁾

\mathcal{H}_a is the part of the dipole-dipole Hamiltonian that is secular relative to \mathcal{H}_s . \mathcal{H}_L is the Hamiltonian of the lattice, to which are added spin-Hamiltonian terms not containing variables relating to the equidistant part of the spectrum; this Hamiltonian plays the role of a thermostat. \mathcal{H}_{sL} is the spin-lattice interaction which, along with \mathcal{H}_L we shall not give in explicit form.

The interaction with the not-strictly-resonant pump of frequency $\Delta \approx k \Delta_1$ (sk is an integer) can be expressed in terms of P_{mm}^j in the form

$$\mathcal{H}_p^i = \hbar \omega_1 \sum_{j,m=-S+k}^{m=S} a_m (P_{m,m-k}^j + P_{m-k,m}^j) \cos \Delta t, \quad (4)$$

where ω_1 is the Larmor frequency, corresponding to the maximum amplitude of the microwave field, a_m are dimensionless quantities, while the nonresonant interaction with an ultrasonic wave of frequency Ω propagating along the z axis can be expressed in the form

$$\mathcal{H}_n^i = \hbar G e_0 \sum_j F_m P_{mm}^j \cos \Omega (t - v^{-1} z_j), \quad (5)$$

where $\hbar G$ is the spin-phonon coupling constant, F_m are dimensionless coefficients, and z_j is the z coordinate of the j -th spin.

The macroscopic state of the system is characterized by nonequilibrium mean values of \mathcal{H}_s and \mathcal{H}_d , which are set in correspondence with the reciprocal temperatures $\beta_s = 1/k_B T_s$, $\beta_d = 1/k_B T_{dd}$. The quantity $\beta_0 = 1/k_B T$ corresponds to the thermostat. The kinetic equations for the reciprocal temperatures are conveniently obtained by the method of the nonequilibrium statistical operator (NSO).⁸ In the case we have considered, when $[\mathcal{H}_s^t, \mathcal{H}_d] \neq 0$,⁶ the absorption of the nonresonant ultrasound is described by the first approximation of the NSO in terms of \mathcal{H}_s^t , while the nonresonant relaxation absorption (or in parallel fields) is determined by the non-vanishing of the commutator $[\mathcal{H}_s^t, \mathcal{H}_{sL}]$ and is contained in the next-order approximation of the NSO.⁹

Averaging the equations of motion of the operators \mathcal{H}_α ($\alpha = s, d$) with the NSO, we arrive in the high-temperature approximation at the next kinetic equations:

$$-\frac{d\beta_s}{dt} = W(\Delta) \left[\beta_s + \frac{\Delta - k\Delta_0}{k\Delta_0} \beta_d \right] + \frac{\beta_s - \beta_0}{\tau_{ss}} + \frac{\beta_d - \beta_0}{\tau_{ds}} \frac{\omega_d^2}{\Delta_0^2}, \quad (6)$$

$$-\frac{d\beta_d}{dt} = W(\Delta) \frac{k\Delta_0(\Delta - k\Delta_0)}{k^2\omega_d^2} \left[\beta_s + \frac{\Delta - k\Delta_0}{k\Delta_0} \beta_d \right] + \frac{\beta_d - \beta_0}{\tau_{dd}} + \frac{\beta_s - \beta_0}{\tau_{ds}} + \frac{\beta_d}{\tau_a}, \quad (7)$$

where the parameters of the spin-lattice relaxation

$$\frac{1}{\tau_{\alpha s}} = \frac{1}{2\omega_\alpha^2} \int_{-\infty}^{\infty} \hbar^{-1} \text{Sp} \{ [\mathcal{H}_\alpha(t_1), \mathcal{H}_{sL}(t_1)] [\mathcal{H}_{sL}, \mathcal{H}_\alpha] \} \text{Sp} \{ S_z^2 \exp(-\beta_s \mathcal{H}_s) \} dt_1 \quad (8)$$

are usually of the same order of magnitude, and $\omega_\alpha^2 = \text{Sp}(\mathcal{H}_\alpha^2)/\hbar^2 \text{Sp}(S_z^2)$. In Ref. 8 and everywhere below, the time dependence of the operators is determined by the interaction representation with the Hamiltonian \mathcal{H}_0 . Finally,

$$W(\Delta) = \frac{3\omega_s^2 k^2 \pi f(\Delta - k\Delta_0)}{S(S+1)(2S+1)} \sum_{m=-S+1}^S a_m^2 \quad (9)$$

is the probability per unit time of transition, within the equidistant part of the spectrum, under the action of not-strictly-resonant pumping, and $f(\Delta - k\Delta_0)$ is the shape of the line of the corresponding transition.

Equations (6) and (7) differ in form from the equations of Provotorov by the term β_d/τ_a in the right hand side of (7). The following expression is obtained for the quantity $1/\tau_a$, which characterizes the degree of the action of the ultrasound on the DDR temperature:

$$\frac{1}{\tau} = \frac{1}{4} G^2 \varepsilon_0^2 \omega_d^{-2} \{ \text{Sp}(S_z^2) \}^{-1} \left\{ \int_{-\infty}^{\infty} \cos \Omega t_1 [\text{Sp}(AA(t_1)) + \sum_{ij} 2 \sin^2 \left(\frac{\Omega}{v} z_{ij} \right) \text{Sp}(A_i A_j(t_1))] dt_1 + \int_{-\infty}^{\infty} \sin \Omega t_1 \left[\sum_{ij} \sin \left(\frac{\Omega}{v} z_{ij} \right) \times \text{Sp}(A_i A_j(t_1))] dt_1 \right\}, \quad (10)$$

where

$$A = \sum_j A_j = \sum_j \left(\sum_m F_m P_{mm}^j \right), \quad A = -\frac{i}{\hbar} [A, \mathcal{H}_0], \quad z_{ij} = z_i - z_j.$$

For a two-level system, as is easily seen, the operator $\dot{A} = 0$, and the effect of absorption is very weak, since the quantity $(\Omega z_{ij}^2/v) \leq 10^{-5}$ for ultrasound. For many-level systems (except for the case $A \sim S_z$), $A \neq 0$ and the corresponding term in (10) is the leading one. After integrating twice by parts in (10), we obtain

$$\frac{1}{\tau_a} = \frac{\pi}{2} G^2 \varepsilon_0^2 \frac{\text{Sp}(\bar{A}^2) \Omega^2}{\text{Sp}(S_z^2) \omega_d^2} g(\Omega) = \varepsilon_0^2 \frac{1}{\tau_a(1)}, \quad (11)$$

where

$$\bar{A} = A - \frac{\text{Sp}(AS_z)}{\text{Sp}(S_z^2)} S_z$$

is the part of A that does not commute with \mathcal{H}_d , and

$$g(\Omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \cos(\Omega t_1) \frac{\text{Sp}[\bar{A}(t_1)\bar{A}]}{\text{Sp}(\bar{A}^2)} dt_1. \quad (12)$$

The ultrasonic absorption coefficient

$$\alpha = \frac{\partial \langle \mathcal{H}_d \rangle}{\partial t} \Big|_a / IV,$$

is measured experimentally. Here $I = \rho v^3 \varepsilon_0^2 / 2$ is the energy passing per unit time through a unit area perpendicular to the direction of the sound wave, and V is the volume of the sample.

If the ultrasonic field is sufficiently weak, so that we can neglect any change in the DDR temperature under its action, then α is found to be the same as in Ref. 6:

$$\alpha = \frac{1}{3} \hbar^2 S(S+1) \frac{2S+1}{n} \frac{N}{V} \beta_d^{st}(0) \omega_d^2 \tau_a^{-1}(1) / \frac{1}{2} \rho v^3, \quad (13)$$

where N is the number of spins in the sample, and $\beta_\alpha^{st}(0)$ is a stationary solution of Eqs. (6) and (7) if we set $\tau_a^{-1} = 0$ in them, i. e., the solution of the Provotorov equations.

In the case $|\delta| = |\Delta - k\Delta_0| \ll \Delta_0$, the values of $\beta_\alpha^{st}(0)$ are given by the following expressions:

$$\frac{\beta_s^{st}(0)}{\beta_0} = \left(\delta^2 + k^2 \omega_d^2 \frac{1}{s(\Delta)} \frac{\tau_{ss}}{\tau_{dd}} \right) / \left(\delta^2 + k^2 \omega_d^2 \frac{s(\Delta)+1}{s(\Delta)} \frac{\tau_{ss}}{\tau_{dd}} \right), \quad (14)$$

$$\frac{\beta_d^{st}(0)}{\beta_0} = \left(-k\Delta_0 \delta + k^2 \omega_d^2 \frac{s(\Delta)+1}{s(\Delta)} \frac{\tau_{ss}}{\tau_{dd}} \right) / \left(\delta^2 + k^2 \omega_d^2 \frac{s(\Delta)+1}{s(\Delta)} \frac{\tau_{ss}}{\tau_{dd}} \right),$$

where $(\Delta) = W(\Delta)\tau_{ss}$ is the microwave pumping factor.

As the ultrasonic power increases, the β_α^{st} begin to differ from (14). If the factor $s(\Delta)$ is such that in the absence of ultrasound a significant shift is observed in the DDR temperature ($\beta_d^{st}(0)/\beta_0 \gg 1$), then the stationary solutions of Eqs. (6) and (7) have the form

$$\beta_d^{st} = \beta_d^{st}(0) \left[\frac{s(\Delta)+1}{s(\Delta)} \frac{\tau_{ss}}{\tau_a} \frac{k^2 \omega_d^2}{|\delta|k\Delta_0} \frac{\beta_d^{st}(0)}{\beta_0} + 1 \right]^{-1}, \quad (15)$$

$$\beta_s^{st} = \beta_s^{st}(0) \left[1 + \frac{k^2 \omega_d^2}{\delta^2 + k^2 \omega_d^2 \tau_{ss}/s(\Delta) \tau_{dd}} \frac{1}{s(\Delta)} \frac{\tau_{ss}}{\tau_a} \right] \times \left[1 + \frac{s(\Delta)+1}{s(\Delta)} \frac{\tau_{ss}}{\tau_a} \frac{k^2 \omega_d^2}{\delta^2 + k^2 \omega_d^2 \tau_{ss}/s(\Delta) \tau_{dd}} \frac{\beta_s^{st}(0)}{\beta_0} \right]^{-1}.$$

It is seen from (15) that with increase in the intensity of the ultrasonic wave ($1/\tau_a \sim I$), the value of β_d^{st} , decreases as a consequence of the direct saturation of the DDR by the ultrasound. Moreover, the barrier to saturation of the energy reservoir connected with the single-particle spectrum of the system is removed, and β_s^{st} can also decrease all the way to the value $\beta_s^{st} = \beta_0 / (s(\Delta)+1)$, while in the absence of ultrasound, even at $s(\Delta) \gg 1$, the value of $\beta_s^{st}(0)$ cannot be less than $\delta^2 / (\delta^2 + k^2 \omega_d^2 \tau_{ss}/\tau_{dd})$. [We recall that in strictly resonant pumping, $\beta_s^{st}(\text{res}) = \beta_0 / (s(k\Delta_0) + 1)$].

The interaction of the nonresonant alternating magnetic field with the Cr^{3+} ions, just as the interaction with ultrasound, does not commute with \mathcal{H}_0 , when there are identical energy levels in the spectrum of the ions.

This is connected with the fact that the splitting of the fundamental multiplet is due, along with the Zeeman energy, to the terms of the spin Hamiltonian which are responsible for the "initial" splitting. It follows from what has been said that the energy of the nonresonant alternating magnetic field is absorbed by the DDR. In order to describe this absorption, we must add the term β_d/τ_h to (7), with the kinetic coefficient τ_h^{-1} proportional to the square of the amplitude h of the alternating magnetic field. If this field is sufficiently weak, then the value of the absorption is proportional to the value of β_d^{st} given by formula (15). It was this circumstance which allowed us, in the experiment described in Sec. 2, to determine the quantity $T_{dd}^0/T_{dd} = \beta_d^{st}/\beta_d^{st}(0)$ from the absorption of the energy of the alternating magnetic field. Expressing the quantity $k^2\omega_d^2\tau_a^{-1}(1)$ with the help of (13) in terms of α and I , we obtain, with account of (14):

$$T_{dd}^0/T_{dd} = [1 + I/I_0]^{-1}, \quad (16)$$

where

$$I_0 = \frac{1}{3} \hbar^2 \left(\frac{N}{V} \right) S(S+1) \frac{2S+1}{n} \frac{(k\Delta_0)|\delta|}{\alpha k^2 k_B T} \frac{s(\Delta)}{s(\Delta)+1} \frac{1}{\tau_{ss}}. \quad (17)$$

As has already been pointed out above, formula (16) satisfactorily describes the experimental results if we set $I_0 \approx 2W/\text{cm}^2$.

The quantity $T/T_{dd}^0(W)$ was determined from the nonresonant absorption as a function of the microwave power $W(s(\Delta) \sim W)$. Upon increase of W to the level used in the experiment on DDR saturation by the ultrasonic field, the $T_{dd}(W)$ dependence became very weak. Consequently, a sufficiently strong ($s(\Delta) \gg 1$), not strictly resonant microwave pumping was realized, with the quantity $(s(\Delta)+1)/s(\Delta) \approx 1$. Substituting I_0 in (17), $\alpha = 10^{-7} \text{ cm}^{-1}$, $T = 4.2 \text{ K}$, $S = 1$, $n = 4$, $N/V = 2.3 \cdot 10^{19} \text{ cm}^{-3}$, $\Delta_0 = 2\pi \cdot 4.7 \cdot 10^9 \text{ c}^{-1}$, $|\delta| = 2\pi \cdot 1.1 \cdot 10^8 \text{ sec}^{-1}$, $k = 2$, we obtain $\tau_{ss} \approx 100 \text{ msec}$. The value of τ_{ss} agrees with the data of other experiments.⁷

In conclusion, we note that the possibility of determination, by acoustic nonresonant methods, of the

spin-phonon coupling constant⁶ and the relaxation times, together with the definite experimental advantages of low-frequency methods, obviously make them promising for the investigation of many paramagnetic materials.

¹) By intensity of the ultrasonic field is meant here the quantity $I = \rho v^3 \epsilon_0^2 / 2 = QPv/V\Omega$, where $v = 11.2 \times 10^5 \text{ cm/sec}$ is the velocity of longitudinal ultrasound, ρ and V are the density and volume of the sample, P is the power fed to the sample, and ϵ_0 is the amplitude of the strain created by the ultrasound.

²) The authors are grateful to I. N. Kurkin for help in the measurement of T_1 .

³) The quantities S and S_z should everywhere be taken in the sense in which they have been introduced here and should not be confused with the spin of the paramagnetic ion.

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