ROLE OF SPECTRAL DIFFUSION AND DIPOLE-DIPOLE RESERVOIR IN THE SATURATION OF AN INHOMOGENEOUSLY BROADENED LINE

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A consistent quantum-statistical study is made of the saturation of an inhomogeneously broadened EPR line with allowance for spectral diffusion and the dipole-dipole reservoir. It is shown that the effect of the dipole-dipole reservoir is negligible in the case of inhomogeneous broadening.

1. Experiments on stationary saturation of resonance can be carried out in two ways.

Scheme A. A saturating alternating field is applied, and its frequency is slowly varied (it is more convenient in the experiment to vary the magnitude of the main field); the corresponding signal is then measured as a function of the frequency.

Scheme B. A saturating field of definite frequency is applied. A second non-saturating field is also applied, its frequency is varied, and the corresponding signal is measured.

Let Ω and H_1 denote the frequency and half the amplitude of the saturating field, and let ω and h_1 denote the frequency and half-amplitude of the alternating field, the corresponding signal of which is measured in the experiment. In scheme A we have $\Omega = \omega$ and $H_1 = h_1$, while in scheme B, $\Omega \neq \omega$ and $H_1 \gg h_1$. Let $\chi''(\omega, \Omega)$ denote the imaginary part of the complex magnetic susceptibility for scheme B. The power absorbed by the sample (from the source of the alternating field ω, h_1) is given by the usual formula

$$P(\omega, \Omega) = 2\omega\chi''(\omega, \Omega)h_1^2. \tag{1}$$

It is easy to see that for scheme A we get

$$\chi''(\omega) = \chi''(\omega, \omega). \tag{2}$$

It is well known[1] that there are two types of broadening of the magnetic resonance absorption line—homogeneous and inhomogeneous. The question of the saturation of a homogeneously broadened resonance line was considered in the papers of Bloembergen, Purcell, and Pound[2] and Bloch[3]. In these papers, however, no account was taken of the significant role that the dipole-dipole reservoir (DDR) of the spins can play in solids. The saturation of the resonance of a homogeneously broadened line with allowance for DDR was considered by Redfield[4] and Provotorov[5] in the cases of strong and intermediate saturation, respectively.

In view of the complications arising in the investigation of inhomogeneously broadened resonance lines, a simplification is introduced, namely, it is assumed that different identical spins are in different local fields. Therefore the inhomogeneously broadened line is represented as consisting of narrow spin packets, and the broadening of each packet is assumed to be homogeneous. Individual spin packets in the homogeneously broadened line are not independent, generally speaking, as is the case in the so called spectral diffusion[6,7]. The calculation of $\chi''(\omega)$ for an inhomogeneously broadened line is given in[1,8], and the calculation of $\chi''(\omega, \Omega)$ in[9,10]. In these calculations, however, the DDR and spectral diffusion were not taken into account. The value of $\chi''(\omega, \Omega)$ with allowance for the role of the DDR was calculated in[10,11], but in the latter papers each spin packet was ascribed its own DDR (it will be shown later that this assumption is not always true), and the spectral diffusion was not taken into account. The question of the saturation of an inhomogeneously broadened line with account of the spectral diffusion but without account of the DDR was considered in[6].

The purpose of the present article is to present a theoretical analysis of the stationary saturation of an inhomogeneously broadened resonance line for a spin system with $S = 1/2$ (S is the effective spin) and with allowance for the DDR and the spectral diffusion. We confine ourselves here to the case of intermediate saturation (the amplitudes of the alternating field is assumed smaller than the width of the spin packet).

2. For the static susceptibility χ_0 of the spin system we have

$$\chi_0 = {}^1/_4 N\gamma^2\beta_L, \tag{3}$$

where N is the spin concentration, γ is the gyromagnetic ratio, β_L is the reciprocal lattice temperature in energy units (we assume $\hbar = 1$ and $S = 1/2$). We denote by $\varphi(\omega - \omega')$ and $g(\omega - \omega')$ the normalized form functions of the spin packet with frequency ω' and of the inhomogeneous line with central frequency $\omega_0 = \gamma H$ (H—constant magnetic field). The widths (in frequency units) due to the homogeneous and inhomogeneous broadening will be denoted respectively Δ and Δ^*. In the case of an inhomogeneously broadened line, $\Delta^* > \Delta$. It is frequently assumed that $\varphi(x)$ has a Lorentz form and $g(x)$ a Gaussian form:

$$\varphi(x) = \frac{\Delta}{\pi(x^2 + \Delta^2)}, \quad g(x) = \frac{1}{(2\pi\Delta^{*2})^{1/2}}\exp\left\{-\frac{x^2}{2\Delta^{*2}}\right\}. \tag{4}$$

According to Bloembergen, Purcell, and Pound[2], we have for the probability (per unit time) of spin reorientation under the influence of the alternating field ω, h_1,

$$W(\omega - \omega') = {}^1/_2\pi(\gamma h_1)^2\varphi(\omega - \omega'). \tag{5}$$

We introduce, finally, the symbol $s = (\gamma H_1)^2 T_{SL} T_2$, where $T_2 = 1/\Delta$ and T_{SL} is the spin-lattice relaxation time. For a homogeneously broadened line, s is the saturation parameter at exact resonance. On the other hand, for an inhomogeneously broadened line with independent packets (i.e., in the absence of spectral diffusion), s is the saturation parameter of that packet whose center coincides with the frequency of the alternating field.

3. This raises the question of the method of describing the quasiequilibrium state of the system considered by us (lattice + spin system + alternating external fields). Exchange of Zeeman energy between the spins belonging to one spin packet is much more rapid than between spins belonging to different packets. In this connection, besides the lattice and the alternating field, the Zeeman degrees of freedom of each of the packets can be regarded as individual subsystems characterized by their own thermodynamic parameters.

A particular analysis should be made of the secular part of the dipole-dipole interaction. We shall assume for the time being that the distribution of the spin packets is discrete, and use the notation of[12]. We consider two spin packets, n and n′. The dipole-dipole Hamiltonian (secular part) of the individual packet is given by

$$\mathcal{H}_d^{\alpha}=\frac{1}{2}\sum_{ij}A_{ij}S_{\alpha i}{}^zS_{\alpha j}{}^z+\sum_{ij}B_{ij}S_{\alpha i}{}^+S_{\alpha j}{}^-,\qquad \alpha=n,n',$$

and the interaction between the packets is described by the expression

$$\mathcal{H}'=\mathcal{H}_d^{nn'}+\mathcal{H}_{cr}^{nn'},$$

where

$$\mathcal{H}_d^{nn'}=\frac{1}{2}\sum_{ij}A_{ij}S_{ni}^zS_{n'j}^z,\qquad \mathcal{H}_{cr}^{nn'}=\sum_{ij}B_{ij}S_{ni}^+S_{n'j}^-.$$

$\mathcal{H}_d^{nn'}$ commutes with the Zeeman energy of the packets, but does not commute with $\mathcal{H}_d^a$. Therefore $\mathcal{H}_d^{nn'}$ gives rise to energy exchange between the DDR of the individual packets without changing their Zeeman energy. The indicated exchange is realized (as is also within a single packet) in the first order of perturbation theory. Two possibilities can be considered.

A. The case when the frequency distribution of the packets corresponds to the spatial distribution of the spin, i.e., closely located spins have close Larmor frequencies (as is the case for inhomogeneous broadening due, for example, to the inhomogeneity of the constant magnetic field). Then the interaction between the spins of the same packet will be stronger than the interaction between the spins of different packets. In such a case, the DDR of an individual packet should be regarded as a separate subsystem, leading to the analysis given in[10,11].

B. In most cases, particularly for the most important and interesting case of homogeneous broadening due to the hyperfine interaction of the spin of the magnetic ion with the spins of the surrounding nuclei[1)] (alkali-halide crystal with F centers, semiconductor with donor or acceptor impurity, diluted paramagnetic salt), the frequency distribution of the spins does not correlate with their spatial distribution. Then the interaction between spins of different packets is of the same order as the interaction between spins of one packet. The energy exchange between the DDR of different packets occurs at the same rate as exchange of energy within one packet, and it is therefore necessary to introduce a single DDR for the entire spin system. We confine ourselves to this case only.

In view of the statements made above, we introduce for the description of the quasiequilibrium state of our system the reciprocal temperatures (in energy units) of the lattice (β_L), of the Zeeman degrees of freedom of the spin packet with central frequency ω, ($\beta(\Omega,\omega')$, and of the DDR ($\beta_d(\Omega)$) (the alternating field is regarded as a subsystem with $\beta_h=0$[13]).

The stationary equations for $\beta(\Omega,\omega')$ and $\beta_d(\Omega)$ can be obtained from (8) of[12], neglecting in the latter the nuclear terms. In the case of diffusion relaxation and continuous distribution of the spin packets, we obtain

$$\frac{\beta(\Omega,\omega')-\beta_L}{T_{SL}}+2W(\Omega-\omega')\left[\beta(\Omega,\omega')+\frac{\Omega-\omega'}{\omega'}\beta_d(\Omega)\right]$$
$$-\frac{1}{\omega'}\int g(\omega''-\omega_0)W_{cr}(\omega''-\omega')[\omega''\beta(\Omega,\omega'')$$
$$-\omega'\beta(\Omega,\omega')+(\omega'-\omega'')\beta_d(\Omega)]\,d\omega''=0, \tag{5a}$$
$$\frac{\beta_d(\Omega)-\beta_L}{T_{dL}}+\frac{2}{\omega_d^2}\int(\Omega-\omega')g(\omega'-\omega_0)W(\Omega-\omega')[\omega'\beta(\Omega,\omega')$$
$$+(\Omega-\omega')\beta_d(\Omega)]\,d\omega'+\frac{1}{\omega_d^2}\int\omega' g(\omega'-\omega_0)[\beta_d(\Omega)-\beta(\Omega,\omega')]\,d\omega'$$
$$\times\int(\omega'-\omega'')g(\omega''-\omega_0)W_{cr}(\omega'-\omega'')\,d\omega''=0, \tag{5b}$$

where $T_{dL}=(\omega_d^2/\Delta^2)T_{SL}$ is the DDR relaxation time,

$$\omega_d^2=(\mathrm{Sp}\,\mathcal{H}_d^2)\Big/\mathrm{Sp}\Big(\sum_{in}S_{in}{}^z\Big)^2$$

plays the role of the square of the average quantum energy in the DDR (n—number of packets, i—number of lattice sites), the integrals over the frequencies are taken from $-\infty$ to $+\infty$, $W_{cr}(\omega'-\omega'')\equiv W_{cr}\varphi_{cr}(\omega'-\omega'')$ is the cross relaxation probability (expressions for W_{cr} and $\varphi_{cr}(x)$ are given in[12]).

4. In the high-temperature approximation we have for the excess of the number of spins in the lower state ω' of the packet

$$n(\Omega,\omega')={}^1\!/_2N\omega' g(\omega'-\omega_0)\beta(\Omega,\omega'). \tag{6}$$

Further,

$$P(\omega,\Omega)=-\frac{1}{2}\int\omega\left.\frac{dn(\Omega,\omega')}{dt}\right|_{h_1}d\omega', \tag{7}$$

where $d/dt\,(\;)|_{h_1}$ denotes the change due to the alternating field ω, h_1. Taking the DDR into account, we obtain[5]

[1)]At first glance it might appear that it is meaningless to introduce the concept of the spin packet in this case. Indeed, under ordinary conditions, the time during which the flip-flop reorientation of a pair of neighboring nuclei takes place is of the order of $10^{-2}-10^{-4}$ sec, and it might be assumed that within such a time the local field at the electron spin will change appreciably. In fact, however, the situation is as follows. The local field remains practically unchanged in the case of a flip-flop transition of the pair of nuclei located sufficiently far from the electron spin. Nor does the local field (more accurately, its component in the external fields, which is yellow when it plays any role) change in a flip-flop transition of a pair of nuclei located in the same coordination sphere around the electron spin. The local field changes appreciably in the flip-flop transition of a pair of nuclei close to the electron spin, but at different distances from it, but such transitions are difficult (the nuclei have displaced Larmor frequencies).

$$\left.\frac{d\beta(\Omega,\omega')}{dt}\right|_{h_1}=-2W(\omega-\omega')\left[\beta(\Omega,\omega')+\frac{\omega-\omega'}{\omega'}\beta_d(\Omega)\right].\quad(8)$$

Using (1), (3), and (6)–(8) we obtain

$$\chi''(\omega,\Omega)=\frac{\pi}{2}\frac{\chi_0}{\beta_L}\int\omega' g(\omega'-\omega_0)\varphi(\omega'-\omega)\Big[\beta(\Omega,\omega')+\frac{\omega-\omega'}{\omega'}\beta_d(\Omega)\Big]d\omega'.\quad(9)$$

If the DDR is disregarded, it is necessary to put $\beta_d = 0$ in (9). In the absence of saturation, recognizing that for an inhomogeneously broadened line $g(x)$ varies much more slowly than $\varphi(x)$, we can readily obtain[2)]

$$\chi_0''(\omega,\Omega)=\chi_0''(\omega)={}^1\!/_2\pi\chi_0\omega_0 g(\omega-\omega_0).\quad(10)$$

5. We first consider the simplest case: the packets are independent and the DDR is disregarded. The corresponding value of $\chi''(\omega,\Omega)$ will be denoted by $\chi''_{00}(\omega,\Omega)$. It is easy to obtain[10] (we assume that $\Delta^*\gg\Delta$)

$$\chi_{00}''(\omega,\Omega)=\chi_0''(\omega)F(\omega-\Omega,s),\quad(11)$$

where

$$F(x,s)=\int\frac{\varphi(x+y)\,dy}{1+\pi sT_2^{-1}\varphi(y)}.\quad(12)$$

for a Lorentzian form of $\varphi(x)$ we have

$$F(x,s)=\frac{\xi^4+\xi^2(4+s-s/\sqrt{1+s})+s^2/\sqrt{1+s}}{\xi^4+\xi^2(4+2s)+s^2},\quad(13)$$
$$\xi=T_2|x|=|x|/\Delta.$$

Using the fundamental properties of the function $F(x,s)$, given in[10], we find that when $s\gg1$ the function $\chi''_{00}(\omega)$ decreases greatly compared with its unsaturated value $\chi'_0(\omega)$ in an interval of $|\omega-\Omega|$ of the order of $\sqrt{s}/T_2$. Outside this interval, $\chi''_{00}(\omega,\Omega)$ is approximately equal to its unsaturated value (10). We thus obtain for the width of the hole burnt out upon saturation of the inhomogeneously broadened line[3)]

$$\tilde\Delta=\Delta\sqrt{s+1}.\quad(14)$$

It is easy to see that the width and the depth of the hole increase with increasing s. It is clear that in order for the foregoing results to be valid it is necessary to satisfy the condition $\tilde\Delta\ll\Delta^*$.

6. We shall again neglect the role of the DDR, but take the spectral diffusion into account. Equations (10) then reduce to

$$\frac{\beta(\Omega,\omega')-\beta_L}{T_{SL}}+2W(\Omega-\omega')\beta(\Omega,\omega')-\frac{1}{\omega'}\int g(\omega''-\omega_0)\times W_{cr}(\omega''-\omega')[\omega''\beta(\Omega,\omega'')-\omega'\beta(\Omega,\omega')]\,d\omega''=0.\quad(15)$$

We introduce

$$D(\omega)={}^1\!/_2W_{cr}\Delta_{cr}^2g(\omega-\omega_0),\qquad k=(DT_{SL})^{-1/2}.\quad(16)$$

D is the coefficient of spectral diffusion, $1/k$ is the frequency interval to which the spin excitation is extended as the result of the spectral diffusion within a time T_{SL}, $\Delta^2_{cr}=\int x^2\varphi_{cr}(x)dx$ is the cross-relaxation second moment. In the case $k\Delta>1$, the spectral diffusion is insignificant and the results of Sec. 5 are valid. The most interesting case is

$$\Delta\ll1/k\ll\Delta^*.\quad(17)$$

In other words, the spectral diffusion extends over many spin packets (their number is of the order of $1/k\Delta$), but does not extend over the entire inhomogeneously broadened line.

Equation (15) with the conditions (17) satisfied was solved by us in[12]. We have

$$\frac{\beta(\Omega,\omega)}{\beta_L}=1-\frac{s'}{s'+1}e^{-k|\omega-\Omega|},\quad(18)$$

where $s'=(1/2)\pi(k\Delta)s\ll s$. Thus, the packets located in an interval of $|\omega-\Omega|$ of the order of $1/k$ are saturated to approximately the same degree, while the packets outside this interval are practically not saturated. The role of the saturation parameter (for the saturated packets) is played by the quantity s'; it is of the order of s divided by the number of saturable packets.

We denote the quantity $\chi''(\omega,\Omega)$ for the case under consideration by $\chi''_{d0}(\omega,\Omega)$. According to (9) we have

$$\chi_{d0}''(\omega,\Omega)=\frac{\pi}{2}\chi_0\omega_0\int g(\omega'-\omega_0)\varphi(\omega'-\omega)\frac{\beta(\Omega,\omega')}{\beta_L}d\omega'.\quad(19)$$

the function $\beta(\Omega,\omega')$ changes appreciably over an interval of the order of $1/k$, $\varphi(\omega'-\omega)$ changes appreciably over an interval of the order of Δ, and $g(\omega'-\omega_0)$ over an interval of the order of Δ^*. Taking (17) and (18) into account, we obtain

$$\chi_{d0}''(\omega,\Omega)=\chi_0''(\omega)\Big[1-\frac{s'}{s'+1}e^{-k|\omega-\Omega|}\Big].\quad(20)$$

Thus, in an interval $|\omega-\Omega|$ of the order of $1/k$, the function $\chi''(\omega,\Omega)$ decreases approximately by a factor $s'+1$, and outside this interval it is approximately equal to its unsaturated value. The width of the hole is of the order of $1/k$. Unlike the case $k\Delta>1$, the width of the hole does not change with increasing H_1, and only its depth increases.

The calculation for the case $k\Delta^*\sim1$ is difficult. On the other hand, if $k\Delta^*<1$, i.e., the spectral diffusion extends over the entire inhomogeneous broadened line, the result is simple[14], namely, upon saturation the inhomogeneously broadened line behaves approximately like a homogeneously broadened line (with width Δ^*).

7. For a better understanding of the picture of the saturation of the inhomogeneously broadened line with account taken of the DDR, we recall first the results pertaining to the case of a homogeneously broadened line. According to[5,15] we have

$$\chi''^{\text{hom}}(\omega,\Omega)=\frac{\pi}{2}\chi_0\omega_0\varphi(\omega-\omega_0)\Big[\beta_z(\Omega)+\frac{\omega-\omega_0}{\omega_0}\beta_d(\Omega)\Big]\frac{1}{\beta_L},\quad(21)$$

where $\beta_z(\Omega)$ is the reciprocal Zeeman temperature. The physical meaning of the expression $\omega_0\beta_z+(\omega-\omega_0)\beta_d$ lies in the fact that when a quantum of alternating field ω is absorbed, an energy ω_0 is absorbed by the Zeeman system, and an energy $\omega-\omega_0$ is absorbed by the DDR. In the case of weak saturation, inasmuch as $\beta_d\approx\beta_z\approx\beta_L$, the quantity $(\omega-\omega_0)\beta_d/\omega_0$ can be neglected, and the DDR does not play any role. On the other hand, in the case of noticeable saturation, owing to the small heat capacity of the DDR (this is

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2)We recall that for a homogeneously broadened line in the absence of saturation we have $\chi''_0(\omega)={}^1\!/_2\pi\chi_0\omega_0\varphi(\omega-\omega_0)$.

3)Replacement of s by s + 1 in (14) corrects the result at small values of s.

connected with the fact that $\omega_d \ll \omega_0$), $|\beta_d|$ increases strongly, and this term can no longer be neglected. In the case when $s \gg 1$ and $|\omega_0 - \Omega|$ is not too small, we get

$$\frac{\beta_d(\Omega)}{\beta_L}=\frac{\omega_0(\omega_0-\Omega)}{\alpha\omega_d^2+(\omega_0-\Omega)^2}, \quad \frac{\beta_z(\Omega)}{\beta_L}=\frac{(\omega_0-\Omega)^2}{\alpha\omega_d^2+(\omega_0-\Omega)^2}, \tag{22}$$

where $\alpha = T_{SL}/T_{dL}$. It is usually assumed that $\alpha = 2$ or $\alpha = 3$[5,15]. It is known that $\alpha\omega_d^2 \sim \Delta^2$. Assuming that $|\omega_0 - \Omega| \sim \Delta$, we obtain

$$|\beta_d^{\rm hom}|/\beta_L \sim \omega_0/\Delta \gg 1. \tag{23}$$

According to (22), $\beta_d(\Omega)$ is negative when $\Omega \gg \omega_0$. This is connected with the fact that in this case the DDR absorbs the quanta $\Omega - \omega_0$; therefore the DDR becomes strongly heated and goes over into the region of negative temperatures.

Formula (21), with (22) taken into account, can be reduced to the form

$$\chi''^{\,\rm hom}(\omega,\Omega)=\frac{\pi}{2}\chi_0\omega_0\varphi(\omega-\omega_0)\frac{(\omega-\Omega)(\omega_0-\Omega)}{\alpha\omega_d^2+(\omega_0-\Omega)^2}. \tag{24}$$

Thus, χ'' is negative, i.e., the maser effect takes place if $\omega > \Omega > \omega_0$ or $\omega < \Omega < \omega_0$.

We note that if $\alpha \gg 1$ an increase of the effective resonance width takes place upon saturation, and the picture becomes similar to that obtained in accordance with Bloembergen, Purcell, and Pound[2]. Since, however, α is of the order of unity, the absorption line will actually become narrower upon saturation.

We proceed to consider the saturation of an inhomogeneously broadened line with allowance for the DDR, but in the absence of spectral diffusion. Equations (5) take the form

$$\frac{\beta(\Omega,\omega')-\beta_L}{T_{SL}}+2W(\Omega-\omega')\left[\beta(\Omega,\omega')+\frac{\Omega-\omega'}{\omega'}\beta_d(\Omega)\right]=0, \tag{25a}$$

$$\frac{\beta_d(\Omega)-\beta_L}{T_{dL}}+\frac{2}{\omega_d^2}\int(\Omega-\omega')g(\omega'-\omega_0)W(\omega'-\Omega)[\omega'\beta(\Omega,\omega') + (\Omega-\omega')\beta_d(\Omega)]d\omega'=0. \tag{25b}$$

The solution of the system (25) is

$$\beta(\Omega,\omega')=\left[\beta_L+\frac{\omega'-\Omega}{\omega'}\frac{\pi s}{T_2}\varphi(\Omega-\omega')\beta_d(\Omega)\right]\left[1+\frac{\pi s}{T_2}\varphi(\Omega-\omega')\right]^{-1}, \tag{26}$$

$$\frac{\beta_d(\Omega)}{\beta_L}=\frac{\alpha\omega_d^2+\Omega\Phi_1(\Omega-\omega_0,s)}{\alpha\omega_d^2+\Phi_2(\Omega-\omega_0,s)}, \tag{27}$$

where

$$\Phi_k(x,s)=\frac{\pi s}{T_2}\int\frac{y^k\varphi(y)g(x+y)}{1+\pi sT_2^{-1}\varphi(y)}dy. \tag{28}$$

In order for (27) to be valid it is necessary to satisfy the condition

$$\omega_0|\Phi_1(\Omega-\omega_0,s)|\gg\Phi_2(\Omega-\omega_0,s). \tag{29}$$

Recognizing that $\Delta^* \gg \Delta$, we can perform an approximate calculation of Φ_1 and Φ_2 (we assume a Lorentz form for $\varphi(y)$; in the calculation of Φ_2 it is assumed, in addition, that $g(y)$ has a Gaussian form). We obtain

$$\Phi_2(x,s)=\frac{s}{s+1}\tilde\Delta^2, \quad \Phi_1(x,s)=-\frac{s}{s+1}\pi^{1/2}\tilde\Delta^2 g(x)\Psi\left(\frac{x}{\sqrt{2}\Delta^*}\right), \tag{30}$$

where

$$\Psi(z)=\int_0^z e^{y^2}dy. \tag{31}$$

Taking (30) into account, Eq. (29) takes the form

$$|\Omega-\omega_0|\gg\Delta^{*2}/\omega_0.$$

Since $\alpha\omega_d^2 \sim \Delta^2$, we find that when $s \gg 1$ it is possible to neglect $\alpha\omega_d^2$ both in the numerator and in the denominator of (27). This yields

$$\frac{\beta_d(\Omega)}{\beta_L}=\frac{\omega_0\Phi_1(\Omega-\omega_0,s)}{\Phi_2(\Omega-\omega_0,s)}=-\pi^{1/2}\omega_0 g(\Omega-\omega_0)\Psi\left(\frac{x}{\sqrt{2}\Delta^*}\right). \tag{32}$$

Thus

$$\frac{|\beta_d(\Omega)|}{\beta_L}\sim\begin{cases}\omega_0/\Delta^*\gg1 & \text{for } |\Omega-\omega_0|\sim\Delta^*\\ \omega_0\tilde\Delta/\Delta^{*2}\gg1 & \text{for } |\Omega-\omega_0|\sim\tilde\Delta\end{cases} \tag{33}$$

Comparison of (33) with (23) shows that in the case of an inhomogeneously broadened line $|\beta_d(\Omega)|$ is much smaller than in the case of homogeneous broadening. This result is connected with the fact that there is one common DDR, and therefore its specific heat is sufficiently large (roughly speaking, this is equivalent to replacing $\alpha\omega_d^2$ in (22) by $(\Delta^*/\Delta)\alpha\omega_d^2$, and then when $|\Omega - \omega_0| \sim \Delta^*$ we obtain $|\beta_d(\Omega)|/\beta_L \sim \omega_0/\Delta^*$ in accordance with (33)).

We denote the quantity $\chi''(\omega, \Omega)$ in the absence of spectral diffusion, but with DDR taken into account, by $\chi''_{0d}(\omega,\Omega)$. When $\Omega = \omega$ we readily obtain with the aid of (32)

$$\chi_{0d}''(\omega)=\chi_{00}''(\omega)\left[1-\tilde\Delta g(\omega-\omega_0)\Psi^2\left(\frac{x}{\sqrt{2}\Delta^*}\right)\right]. \tag{34}$$

The second term in the square brackets is always small compared with unity, and therefore $\chi''_{0d}(\omega) \approx \chi''_{00}(\omega)$.

We can also calculate $\chi''_{0d}(\omega, \Omega)$ with the condition $|\omega - \Omega| > \tilde\Delta$ satisfied. We obtain

$$\chi_{0d}''(\omega,\Omega)=\chi_{00}''(\omega,\Omega)\left\{1-\frac{\beta_d(\Omega)}{\beta_L}\frac{\omega-\Omega}{\omega_0}[1-F^{-1}(\omega-\Omega,s)]\right\}. \tag{35}$$

A simple estimate shows[4)] that the second term in the curly brackets of (35) is always much smaller than unity. It can thus be regarded as established that

$$\chi_{0d}''(\omega,\Omega)\approx\chi_{00}''(\omega,\Omega) \tag{36}$$

for any relation between ω and Ω.

Thus, the role of the DDR in the saturation of an inhomogeneous line is negligible in the case of independent packets. Physically this result can be understood as follows. Since the width $\tilde\Delta$ of the saturated packet subtends many Zeeman frequencies, the energy acquired or given up by the DDR is determined completely by the asymmetry of the distribution of the Zeeman frequencies relative to the frequency of the saturating field. This asymmetry is characterized by the parameter

$$\left|\frac{dg(\omega-\omega_0)}{d\omega}\right|\tilde\Delta\sim\frac{|\omega-\omega_0|\tilde\Delta}{\Delta^{*2}}\sim\frac{\tilde\Delta}{\Delta^*}$$

and leads to the appearance of an additional small factor $\tilde\Delta/\Delta^*$ in the second term of (21), as the result of which the DDR ceases to play an appreciable role.

9. Let us consider, finally, the question of the saturation of the inhomogeneously broadened line with

4)In the estimate of the order of magnitude it is possible to use formula (35) also in the case when $|\omega - \Omega| \sim \tilde\Delta$.

allowance of the DDR in the case when the spectral diffusion is appreciable, i.e., $k\Delta < 1$. It is easy to see that when the conditions (17) are satisfied the role of the width of the hole is played not by Δ but by $1/k$, and asymmetry parameter is of the order of $1/k\Delta^* \ll 1$. Consequently, the DDR will not play any role, and the results obtained in Sec. 7 will remain in force. As to the quantity β_d, on the basis of formula (22) and of the remark following Eq. (33) concerning the specific heat of the DDR, it is easy to obtain the following qualitative estimate:

$$\frac{\beta_d}{\beta_L} \sim \frac{k\omega_0}{(k\Delta)^2 k\Delta^* + 1}. \tag{37}$$

On the other hand, if the condition $k\Delta^* < 1$ is satisfied, then the asymmetry parameter will be of the order of unity and $\beta_d/\beta_L \sim \omega_0/\Delta^*$, i.e., we can expect the situation to be analogous to that obtaining in the saturation of a homogeneous line with a width equal to Δ^*.

10. It follows from all the foregoing that in the case of inhomogeneous broadening it is difficult to observe the DDR temperature change with the aid of saturation of the EPR absorption line. On the other hand, if $\omega_d \sim \omega_I$ (ω_I is the NMR frequency), then the nuclei relax to the electronic DDR, and by virtue of (37) nuclear polarization is possible in the case of EPR saturation. This polarization greatly influences the value of the NMR, making it possible to observe the change of the DDR temperature in the inhomogeneous case.

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