

As a result,

$$I = \ln \frac{8\omega_s \tau_L}{81} - 9 - 2\psi\left(\rho + \frac{1}{2}\right) - 2\psi\left(\rho - \frac{1}{2}\right) - i \frac{\pi}{2}, \quad (\text{A. 24})$$

which transforms into (3. 19) at  $\rho = \frac{1}{6}$ .

- <sup>1</sup>The velocity  $c_0$  is a variable quantity: it depends on the density  $\rho$ . The definition (2. 7), however, contains the constant quantity  $c_0(\rho_0)$ , where  $\rho_0$  is the mean density of HeII. With such a definition, small departures  $\delta\rho_n$  from the equilibrium value of  $\rho_n$  are proportional to the small departure  $\delta s$  from the equilibrium value of the entropy density  $S$ , and  $\delta\rho_n/\rho_n = (4/3)\delta S/S$ .
- <sup>2</sup>The deviation of the phonon spectrum from linear is taken into account only in the calculation of the transverse relaxation operator for the phonons.
- <sup>3</sup>In particular, the last term in (2. 19) can differ from zero for this reason. In this case, we mean by  $\tau_{ij}$  a constant quantity (in contrast, for example, to the nonlinear term in Eq. (18. 7), of Ref. 18, where  $\tau_{ij}$  denotes the complete viscous stress tensor).
- <sup>4</sup>To make estimates of such a type more precise, we must keep it in mind that the characteristic value  $\varepsilon_n$  of the quantity  $\varepsilon(k)$ , which determines the rate of transverse relaxation in the system of phonons, must, as noted in Ref. 12, be of the order of  $\xi(AT/\hbar c)$ , where  $A$  is a number of the order of 10.
- <sup>5</sup>If this condition is violated, then the constant part  $v_n$  can be of the order of  $c$ , which violates the limits of applicability of the initial equations (2. 17)–(2. 19) themselves.

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## The effect of a strong ac field on the dynamical nuclear resonance frequency shift in magnetically ordered crystals

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We use the example of strong stationary NMR saturation in magnetically ordered crystals to show that due to the change in the interaction between spins the magnitude of the dynamical frequency shift (DFS) depends not only on the average value of the  $z$ -component of the nuclear spin, but also on the relation between the detuning and the amplitude of the strong ac field. We show also that for an appropriate choice of the parameters of that field there may be no DFS.

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It is well known<sup>[1,2]</sup> that the hyperfine interaction between nuclear and electron spins in magnetically ordered materials leads to the occurrence of a strong indirect interaction between the nuclear spins (Suhl–Nakamura interaction<sup>[3]</sup>). This interaction determines the first and second moments of the NMR line. We have  $M_1 \gg \sqrt{M_2}$  even at high temperatures ( $\hbar\omega/kT \ll 1$ ).<sup>[4]</sup> The first moment of the line or, alternatively, the dynamical frequency shift (DFS) depends linearly on the aver-

age value of the  $z$ -component of the nuclear spin (in temperature terms: on the reciprocal temperature of the nuclear spin system). This fact leads to a change in the resonance frequency for NMR saturation.

So far<sup>[5,6]</sup> the effect of a strong ac field on the magnitude of the DFS has been considered solely in terms of the change in the  $z$ -component of the magnetization. However, if the ac field is sufficiently large, it changes

appreciably also the magnitude of the interaction itself which determines the first line moment. For instance, <sup>[7]</sup> if we have the "magic angle" situation, the interaction vanishes in first approximation, i. e., we may expect that for an appropriate choice of the ac field parameters the magnitude of the DFS is changed considerably.

We show this in the present paper, using as an example strong stationary saturation.

Following <sup>[8]</sup>, we consider a nuclear spin system on which are acting two rotating magnetic fields: one a saturating one with amplitude  $H_{1p}$  and frequency  $\omega_p$  and the other a detecting one with amplitude  $H_{1d}$  and frequency  $\omega_d$ . We write the Hamiltonian of the system considered in the form

$$\mathcal{H} = \mathcal{H}_I + \mathcal{H}_{SN} + \mathcal{H}_{Ip} + \mathcal{H}_{Id},$$

$$\mathcal{H}_I = \omega_I I^z,$$

$$\mathcal{H}_{SN} = \sum_{i,j} u_{ij} I_i^+ I_j^- \quad I^\pm = I^x \pm iI^y,$$

$$\mathcal{H}_{Ip,d} = \frac{1}{2} \omega_{Ip,d} \{ I^+ \exp(-i\omega_p t) + I^- \exp(i\omega_p t) \},$$

$$\omega_{Ip,d} = \gamma H_{Ip,d},$$

where  $\mathcal{H}_I$  and  $\omega_I$  are the Zeeman energy and frequency of the nuclear spins,  $I^{x,y,z}$  the nuclear spin operator,  $\mathcal{H}_{SN}$  the Suhl-Nakamura interaction Hamiltonian, and  $\mathcal{H}_{Ip,d}$  the Hamiltonians for the interaction of the spin system with the ac fields.

Considering  $\mathcal{H}_{Id}$  to be a small perturbation we can obtain an expression for the power absorbed from the ac field of frequency  $\omega_d$ :

$$Q_d = 2\omega_d H_{1d}^2 \chi''(\omega_p, \omega_d);$$

here

$$\begin{aligned} \chi'' &= \frac{1}{2} \gamma_I^2 \operatorname{Re} \int_0^{\infty} \operatorname{Sp} \{ \rho [I^-(\tau), I^+] \} \exp[i(\omega_d - \omega_p)\tau] d\tau, \\ I^-(\tau) &= \exp(i\mathcal{H}^* \tau) I^- \exp(-i\mathcal{H}^* \tau), \\ \mathcal{H}^* &= (\omega_I - \omega_p) I^z + \omega_{Ip} I^+ + \mathcal{H}_{SN}, \\ \rho &= \exp(-\beta_I \mathcal{H}^*) / \operatorname{Sp} 1, \end{aligned}$$

$\mathcal{H}^*$  is the Hamiltonian of the system in the system of coordinates (RSC) which rotates around the  $z$ -axis with frequency  $\omega_p$ ,  $\beta_I$  is the reciprocal spin temperature of the nuclei in the RSC, and  $\gamma_I$  the gyromagnetic ratio.

Using the canonical transformation

$$\begin{aligned} U &= \exp(i\theta I^y), \\ \sin \theta &= \omega_{Ip} / \omega_s, \quad \cos \theta = (\omega_I - \omega_p) / \omega_s, \\ \omega_s^2 &= (\omega_I - \omega_p)^2 + \omega_{Ip}^2 \end{aligned} \quad (1)$$

we can express  $\chi''$  in the following form:

$$\begin{aligned} \chi'' &= \frac{1}{2} \gamma_I^2 \operatorname{Re} \int_0^{\infty} \{ \Lambda_+^2(\theta) I^+(\tau) + \Lambda_-^2(\theta) I^-(\tau) \} \exp[i(\omega_d - \omega_p)\tau] d\tau, \\ I^{\pm}(\tau) &= \operatorname{Sp} \{ \beta [I^{\pm}(\tau), I^{\mp}(\tau)] \}, \quad \beta = U \rho U^{-1}, \\ I^{\pm}(\tau) &= \exp(i\mathcal{H}^* \tau) I^{\pm} \exp(-i\mathcal{H}^* \tau), \quad \mathcal{H}^* = \omega_s I^z + \lambda(\theta) \mathcal{H}_{SN}, \\ \lambda(\theta) &= \frac{1}{2} (3 \cos^2 \theta - 1), \quad \Lambda_{\pm}^2(\theta) = \frac{1}{2} (\cos \theta \pm 1), \end{aligned} \quad (2)$$

where  $I^{x,y,z}$  are the spin operators in the new system of coordinates.

We retained here in  $\mathcal{H}^*$  that part of the Suhl-Nakamura

interaction which is secular in  $I^z$ . This is necessary for a correct evaluation of the first line moment in the case when  $\omega_{1p}$  is much larger than the DFS. The appearance of the factor  $\lambda(\theta)$  is connected with the change to the new system of coordinates in (1). <sup>[9]</sup>

Writing down the equations of motion for the correlators  $I^{**}(\tau)$  and using the decoupling <sup>[2]</sup>

$$\begin{aligned} \langle [I^+(\tau) I^-, I^-] \rangle &= \langle I^+ \rangle \langle [I^+(\tau), I^-] \rangle, \\ \langle \dots \rangle &= \operatorname{Sp} \{ \beta \dots \} / \operatorname{Sp} 1, \end{aligned}$$

we can obtain

$$\begin{aligned} I^{\pm}(\tau) &= \mp 2\beta_I \omega_s \overline{(I^z)^2} \exp[\pm i(\omega_s - \Delta\omega_s)\tau], \\ \Delta\omega_s &= \frac{2}{N} \beta_I \omega_s \overline{(I^z)^2} \lambda(\theta) \sum_i u_{ii}, \\ \overline{(I^z)^2} &= \operatorname{Sp} (I^z)^2 = \frac{1}{2} NI(I+1). \end{aligned} \quad (3)$$

Substituting (3) into (2) we easily get

$$\chi'' = \frac{1}{2} \gamma_I^2 \omega_s \overline{(I^z)^2} \beta_I \{ \Lambda_+^2(\theta) \delta(\omega_s - \Delta\omega_s - \omega_p + \omega_d) - \Lambda_-^2(\theta) \delta(\omega_s - \Delta\omega_s + \omega_p - \omega_d) \}.$$

The stationary value of  $\beta_I$  in the case when  $\omega_{1p}$  is much larger than the line width or the DFS is given by the formula <sup>[7]</sup>

$$\beta_I \approx \beta_0 \frac{\omega_I(\omega_I - \omega_p) + \omega_{Ip}^2 T_{zL} / T_{xL}}{\omega_{Ip}^2 T_{zL} (1/T'_{xL} + 1/T''_{xL})},$$

where  $T_{zL}$  is the relaxation time of the  $z$ -component of the nuclear spin,  $T'_{xL}$  and  $T''_{xL}$  are the relaxation times of the  $x$ -component of the nuclear spin caused by the secular and the non-secular parts of the spin-lattice interaction in the RSC, and  $\beta_0$  is the equilibrium temperature.

The absorption has thus a resonance character and is observed at the two frequencies  $\omega = \omega_p \pm (\omega_s - \Delta\omega_s)$ ;  $\Delta\omega_s$  is the DFS when the spin system is saturated by the ac field.

The magnitude and sign of the DFS depend on the frequency and amplitude of the saturating field through  $\beta_I$  and  $\lambda(\theta) = (3 \cos^2 \theta - 1)/2$ . In the case when  $\lambda(\theta) = 0$ , i. e.,  $(\omega_I - \omega_p)^2 = \frac{1}{2} \omega_{Ip}^2$ , the DFS will vanish. In that case there will not occur a detuning of the resonance in spin echo experiments. <sup>[5, 6, 10]</sup>

We note also that when  $\cos^2 \theta = \frac{1}{3}$  the DFS changes sign. For instance, if  $\omega_I = \omega_p$ , we have  $\Delta\omega_s = -\frac{1}{2}(\Delta\omega_s)_0$ , where  $(\Delta\omega_s)_0$  is the magnitude of the DFS when  $\omega_{1p} \ll |\omega_I - \omega_p|$ .

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## Optical observation of antiferromagnetic resonance in $\text{CoCO}_3$

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The interaction of light with the spin system during antiferromagnetic resonance (AFMR) was studied in weakly-ferromagnetic  $\text{CoCO}_3$ . The variation of the intensity of the light passing through the optical system, which consisted of a polarizer, the crystal, a compensator, and an analyzer, was measured during excitation of AFMR in the crystal. In order to amplify the weak optical signals, modulation of the microwave power was employed. It was found that under conditions of complete compensation of constant light, the alternating optical signal at resonance was absent. It appears only with discompensation of the transmitted light by means of the compensator. The shape and position of the optical signal coincide with the shape and position of the AFMR signal. The observed optical signal is produced by a change of magnetic birefringence in the crystal; this is due to a decrease, during resonance, of the mean value of the projection of the antiferromagnetic vector on its equilibrium direction.

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### INTRODUCTION

Observation of uniform magnetic resonance by an optical method in magnetically ordered media was first carried out by Dillon on the ferromagnetic crystal  $\text{CrBr}_3$ .<sup>[1]</sup> This method was based on use of the large Faraday effect in the crystals under study and was as follows: on passage of the magnetic field through resonance, the optical system, consisting of the specimen placed between crossed polaroids, cleared because of the appearance of an alternating component of the magnetic moment along the light beam. By means of light it was also possible to observe the spectrum of magnetostatic modes in the specimen.<sup>[1]</sup> Dillon and his co-workers showed theoretically and experimentally, by means of a Fabry-Perot interferometer,<sup>[2]</sup> that the observed optical signal in crossed polaroids occurs at the displaced frequency  $\omega + \Omega$ , where  $\omega$  is the frequency of the incident light and  $\Omega$  is the frequency of the ferromagnetic resonance. Thus in this case there emerges from the system, light modulated by the ferromagnetic-resonance frequency. Later, optical detection of ferromagnetic resonance was carried out by many authors on ferrites (see, for example,<sup>[3]</sup>). But a direct proof of the fact that the observed optical signal in crossed polaroids occurs at the displaced frequency  $\omega + \Omega$  is found only in the spectral experiments of Dillon.<sup>[2]</sup>

We thought it would be interesting to see an optical

signal during excitation of magnetic resonance in antiferromagnetic crystals. We chose as our materials the well-studied weak ferromagnets  $\text{CoCO}_3$  and  $\text{MnCO}_3$ . For optical detection of antiferromagnetic resonance (AFMR) we made use of the presence of magnetic birefringence in these crystals.<sup>[4]</sup>

The crystals  $\text{CoCO}_3$  and  $\text{MnCO}_3$  are rhombohedral; their crystalline symmetry is described by space group  $\bar{D}_{3d}^6$ . The elementary cell of  $\text{CoCO}_3$  and  $\text{MnCO}_3$  contains two magnetic ions.<sup>[5]</sup> At low temperatures these compounds change to an ordered antiferromagnetic state of the easy-plane type. The magnetic moments of the sublattices are canted and form a spontaneous ferromagnetic moment  $m$ , which lies in the basal plane (111) and is perpendicular to the antiferromagnetic vector  $l$ .<sup>[6]</sup> The value of  $m$  is in  $\text{MnCO}_3$  0.2% and in  $\text{CoCO}_3$  about 5% of the nominal value of the sublattice magnetization. According to the data of<sup>[7]</sup>, in  $\text{CoCO}_3$  in the absence of an external magnetic field the vector  $l$  lies in a vertical plane of symmetry at a certain angle (different from  $90^\circ$ ) to the third-order axis. In the basal plane the crystallographic anisotropy of these compounds is small,<sup>[6,8]</sup> and therefore in a magnetic field  $H$  ( $> 0.5$  kOe), regardless of its direction, the vector  $m$  always sets itself along the field.

The AFMR spectrum of the carbonates  $\text{CoCO}_3$  and  $\text{MnCO}_3$  has two branches, which differ significantly with