

# High-temperature spin dynamics and the frequency dependence of the ESR linewidth in low-dimensional Heisenberg magnetic systems

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The frequency dependence of the high-temperature ESR linewidth at the “magic” angle is studied in a number of low-dimensional magnetic systems. The exchange interaction parameters, which determine the coefficient of spin diffusion in these systems, and the single-ion anisotropy parameter agree with the values obtained by other methods of study. The one-dimensional system  $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$  is found to have an anomalously short spin-diffusion cutoff time  $\tau_2 = 5 \cdot 10^{-12}$  sec. The contributions which determine the angular dependence of the linewidth at room temperature are calculated for the two-dimensional system  $\text{BaMnF}_4$ .

## 1. INTRODUCTION

The high-temperature spin dynamics of low-dimensional magnetic insulators having a large spatial anisotropy of the exchange interaction  $J \gg J'$  is described in the models of isolated chains or planes of magnetic ions by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_z + \mathcal{H}' = \mathcal{H}_0 + \mathcal{H}'. \quad (1)$$

Here

$$\mathcal{H}_{\text{ex}} = 2J \sum_{i,j} \mathbf{S}_i \mathbf{S}_j$$

is the isotropic Heisenberg exchange interaction in the chain or plane, and

$$\mathcal{H}_z = g\beta H_0 \sum_n S_n^z$$

is the Zeeman interaction; the broadening interactions subsumed in  $\mathcal{H}'$  include the intrachain or intraplane dipole-dipole interaction, the single-ion anisotropy, the hyperfine interaction, etc.

The theory of magnetic resonance<sup>1</sup> relates the width of an exchange narrowed line to the amplitude of the local fields and, for  $kT \gg J$ , to the Fourier components of the spin correlation functions. The spin correlation function of the system, in turn, is given by

$$\Psi_{KT}(\tau) = \langle [\mathcal{H}'(\tau), M_+] [M_-, \mathcal{H}'] \rangle / \langle M_+ M_- \rangle, \quad (2)$$

where  $M_{\pm} = g\beta \sum_n S_n^{\pm}$  are the components of the total magnetization,  $\langle \dots \rangle$  denotes a temperature average, and the time dependence of  $\mathcal{H}'(\tau)$  is determined by the Hamiltonian  $\mathcal{H}_0$ . The ESR line is the Fourier transform of the spin relaxation function

$$I(\Omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \varphi(t) \exp(-i\Omega t) dt,$$

which is determined by the temporal behavior of  $\Psi(\tau)$ :

$$\varphi(t) = \exp \left[ - \int_0^t (t-\tau) \Psi(\tau) d\tau \right],$$

here  $\Omega = \omega - \omega_0$ , and  $\omega_0 = g\beta H_0 / \hbar$  is the Larmor frequency, which corresponds to the center of the absorption line.

For low-dimensional systems the behavior of the correlation function for  $\tau \rightarrow \infty$  is important, being of a diffusive nature:

$$\Psi_{KT}(\tau) \sim \tau^{-d/2} \quad (3)$$

( $d$  is the dimensionality of the magnetic system). The regime of diffusive behavior of  $\Psi_{KT}(\tau)$  is preceded by a rapid Gaussian correlation dissipation

$$\Psi_{KT}^s(\tau) = M_2 \exp(-\omega_e^2 \tau^2 / 2), \quad (4)$$

which goes over to (3) at  $\tau_1 \sim \hbar / JS$ . Here  $\Psi^s(0) = M_2$  is the second moment of the resonance line, and  $\omega_e \sim J / \hbar$  is the exchange frequency. At long times  $\tau$  there is a cutoff of the diffusion process

$$\Psi(\tau) = \Psi_{KT}(\tau) \exp(-\tau / \tau_2), \quad (5)$$

where  $\tau_2$  is determined, as a rule, by the interchain or interlayer exchange<sup>2,3</sup> or by the intrachain and intralayer dipole-dipole interaction<sup>4,5</sup> and is given in order of magnitude by  $\tau_2 \sim 1 / \gamma \Delta H$ , where  $\Delta H$  is the width of the resonance line.

The total width of the resonance line consists of the short-time and diffusion contributions to  $\Psi(\tau)$ , the latter being dominant for low-dimensional systems, since its spectral density for  $\tau \rightarrow \infty$  has a divergence of the form  $\omega^{-1/2}$  for the one-dimensional case and of the form  $\ln(1/\omega)$  for the two-dimensional case. The main experimental results on the ESR of low-dimensional systems are described in the above model and, as a rule, come from studies of the angular dependence of the width and shape of the absorption line.<sup>2-4,6</sup> The purpose of the present study was to investigate the frequency dependence of the ESR linewidth of low-dimensional magnetic systems and to determine the properties of such systems.

## 2. EXPERIMENTAL TECHNIQUES AND THE PROPERTIES OF THE CRYSTALS

The ESR linewidths were measured in the frequency range  $\sim 10$ –75 GHz on a wide-range spectrometer with a

replaceable waveguide circuit mounted on the base of an SP-47 electromagnet, in fields up to 33 kOe. The design of the cavities allowed the samples to be rotated in two mutually perpendicular planes with respect to the direction of the external field  $H$ . The measurements were made at room temperature, so that  $kT \gg J$ . The absorption lines were usually recorded in the first derivative, whose width  $\Delta H_{pp}$  is related to the familiar width at half maximum by  $\Delta H_{1/2} = 3^{1/2} \Delta H_{pp}$  for Lorentzian lines. The results of previous studies<sup>2,7</sup> of the high-temperature ESR in  $\text{BaMnF}_4$  and  $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$  have shown that near the "magic" angle the shape of the absorption lines is non-Lorentzian only at large distances from the line center  $H_0$ .

To exclude spin-lattice relaxation effects in the study of the high-temperature spin dynamics, we chose low-dimensional compounds of the ion  $\text{Mn}^{2+}$ , which has a purely spin magnetic moment with  $S = 5/2$ . The magnetic and crystallographic properties of the crystals used have been studied rather thoroughly.  $\text{BaMnF}_4$  is an antiferromagnet having a high degree of magnetic two-dimensionality, with  $J'/J \approx 10^{-4}$  (Ref. 8). The interionic distances in the basal plane are  $a/2 = 2.99 \text{ \AA}$  and  $c = 4.22 \text{ \AA}$ , the planes are separated by a distance  $b = 15.1 \text{ \AA}$ , and the magnetic ordering temperature is  $T_N \approx 26 \text{ K}$ . The quasi-one-dimensional antiferromagnet  $[(\text{CH}_3)_3\text{NH}]\text{MnCl}_3 \cdot 2\text{H}_2\text{O}$  (TMAMC) has a structure consisting of chains of  $\text{Mn}^{2+}$  ions with an interionic distance of  $b/2 = 3.71 \text{ \AA}$  and with interchain distances of  $a = 16.733 \text{ \AA}$  and  $c = 8.198 \text{ \AA}$  (Ref. 9). It was noted in Ref. 10 that the single-ion anisotropy has an appreciable contribution of the form  $AS_z^2$ , with an intracrystalline field parameter  $A = 0.1 \text{ cm}^{-1}$ . The Néel temperature of this crystal is  $T_N = 0.98 \text{ K}$ . The quasi-one-dimensional antiferromagnet  $\text{CsMnCl}_3 \cdot 2\text{H}_2\text{O}$  (CMC), with  $T_N = 4.89 \text{ K}$ , consists of chains of magnetic ions with an intrachain separation of  $a/2 = 4.55 \text{ \AA}$  and with interchain distances of  $b = 7.28 \text{ \AA}$  and  $c = 5.75 \text{ \AA}$ . The degree of spatial one-dimensionality in CMC is  $a/2c = 1.3$ , i.e., substantially smaller than in TMAMC, where it has a value of 2.2.

The hyperfine interaction constants of  $\text{Mn}^{2+}$  for ionic crystals are typically less than  $\sim 10^{-2} \text{ cm}^{-1}$ , substantially smaller than the parameters of the other broadening interactions in the systems under study. We have therefore disregarded the hyperfine interaction in the quantitative description of the results.

### 3. DESCRIPTION OF EXPERIMENTAL RESULTS

The spin dipole correlation function (3), including that of low-dimensional systems, can be divided into secular and nonsecular (diagonal and off-diagonal) contributions

$$\Psi(\tau) = \sum_m \Psi_m(\tau) \exp(im\omega_0\tau),$$

where  $m$  is the change in the Zeeman quantum number. The relaxation function in the diffusion limit therefore assumes the form

$$\varphi_d(t) = \exp[-A_1(3 \cos^2 \theta - 1)^2 t^{\frac{1}{2}} - A_2 F_m(\theta) t] \quad (6)$$

for  $d = 1$  (Ref. 11) and

$$\varphi_d(t) = \exp[-B_1(1 - 3 \cos^2 \theta)^2 t \ln t - B_2 F_m(\theta) t] \quad (7)$$

for  $d = 2$  (Ref. 3), where  $\theta$  is the angle between the direction of the external field and the axis of the chain or the normal to the plane of the magnetic ions, and the angular coefficients  $F_m(\theta)$  of the nonsecular terms are calculated from the geometry of the structure.

As a rule, the angular dependence of the ESR linewidth and line shape in low-dimensional magnets is analyzed with allowance for the dominant contribution of the secular terms.<sup>11</sup> However, at the "magic" angle  $\theta_c = \arccos(1/\sqrt{3}) = 54.7^\circ$  these terms vanish, and the nonsecular part and the short time contribution form a Lorentzian line width at  $\theta_c$  is at a minimum and depends on the frequency  $\omega_0$ .

The nonsecular line broadening, which is known in the literature<sup>12</sup> as the " $\frac{10}{3}$  effect," is specific to low-dimensional magnetic systems, since the long-time diffusive behavior of the correlation functions of low-dimensional magnets (3) leads to a definite frequency dependence of the nonsecular linewidth—it is proportional to  $\nu^{-1/2}$  for a one-dimensional system and to  $(-\ln \nu)$  for a  $2d$  system. In fact, with allowance for the concrete form of the angular coefficients  $F_m(\theta)$  the expression for the linewidth of a one-dimensional system at  $\theta_c$  becomes<sup>13</sup>

$$\Delta H_{1/2} = \frac{2}{\gamma} \int_0^\infty \Psi_{dm}(\tau) d\tau = 1.518S(S+1) \frac{g^2 \beta^3 \xi^2(3)}{\hbar l^3} \left( \frac{l^2}{D_1 \nu_0} \right)^{1/2}, \quad (8)$$

where  $\xi(3)$  is the Riemann function,  $D_1$  is the diffusion coefficient,<sup>14</sup> and  $l$  is the interionic distance within the chain.

Figure 1 shows the frequency dependence of the ESR linewidth at the "magic" angle for TMAMC in the coordinates suggested by Eq. (8). The evident linear dependence of  $\Delta H$  on  $\nu_0^{-1/2}$  indicates that this system is described well in the diffusion approximation. When the linear part of the curve is

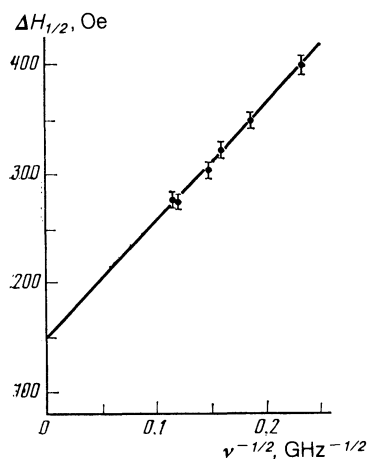


FIG. 1. Frequency dependence of the ESR linewidth  $\Delta H_{pp}$  for  $\theta = \theta_c$  in the  $ab$  plane of the TMAMC crystal at 300 K.

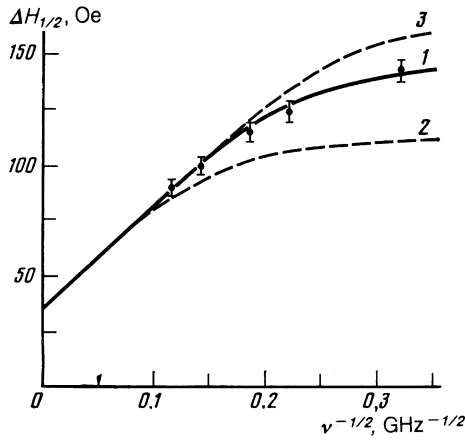


FIG. 2. Frequency dependence of the minimum of the ESR linewidth  $\Delta H_{1/2}$  in the  $ab$  plane of the CMC crystal at  $T = 300$  K. The lines are calculated by Eq. (17) for various values of the parameter  $\tau_2$  (in s): 1)  $5 \cdot 10^{-12}$ ; 2)  $2.5 \cdot 10^{-12}$ ; 3)  $7.5 \cdot 10^{-12}$ .

continued to the region  $\nu \rightarrow \infty$ , it intersects the abscissa at  $\Delta H_0 = 150 \pm 10$  Oe, the value due to nondiffusional contributions to the linewidth.

For CMC, as we see from Fig. 2, the linear dependence of the minimum linewidth is observed only at high frequencies. As the frequency is reduced to  $\sim 15$  GHz, this dependence becomes flatter, indicating that the cutoff mechanism (5) for the one-dimensional diffusion has come into play. The frequency-independent short-time contribution to the linewidth in CMC is  $\Delta H_0 = 35 \pm 10$  Oe. For the low-frequency part of this curve the values of the linewidth agree with data obtained<sup>6</sup> at a frequency of  $\nu_0 = 9.54$  GHz.

A two-dimensional system at  $\theta_c$  should exhibit a frequency-dependent linewidth of the form<sup>3</sup>

$$\Delta H \sim \frac{S(S+1)F_m(\theta)}{D_2} \ln \frac{\omega_e}{\omega_0}. \quad (9)$$

Here  $\omega_e$  is the exchange frequency, and the diffusion coefficient is given by<sup>15</sup>

$$D_2 = \frac{\gamma \sqrt{2\pi} J l^2}{3 \hbar} [S(S+1)]^{1/2}.$$

$\text{BaMnF}_4$  has a linear dependence of  $\Delta H$  on  $\ln \nu_0$  ( $\nu_0 = \omega_0/2\pi$ ) in the investigated frequency range, indicating the diffusion description of the spin dynamics is applicable in this crystal.

#### 4. DISCUSSION OF THE RESULTS

##### a) TMAMC

The angular coefficients of the linear regions of the  $\Delta H(\nu_0)$  curves can be used to determine the parameters of low-dimensional systems. The most important of these, the intrachain or intraplane exchange interaction parameter  $J$ , is a factor in the diffusion coefficient and is the exchange frequency  $\omega_e$ , which determines the value of  $\tau_1$ , i.e., the lower boundary of the diffusion regime (3) in the correlation dissipation. For  $\tau < \tau_1$  the Gaussian correlation dissipation

(4) gives a short-time contribution to the linewidth

$$\Delta H_s = \frac{2}{\gamma} \int_0^\infty \Psi^s(\tau) d\tau = \frac{1}{\gamma} \left( \frac{\pi}{2} \right)^{1/2} \frac{M_2}{\omega_e}, \quad (10)$$

$$M_2 = \frac{\xi(6)}{l^6} \frac{g^4 \beta^4}{\hbar^2} 3S(S+1) (1 + \cos^2 \theta),$$

where  $\gamma = g\beta/\hbar = 1.76 \cdot 10^7$  (Hz·sec)<sup>-1</sup>, and  $\omega_e = 2zJS \times (S+1)$ , with  $z = 2$ .

The diffusion contribution to the linewidth for  $\tau > \tau_1$  is determined by the secular and nonsecular parts of the dipole-dipole interaction

$$\Psi_{dm} = [2F_m(\theta)]^2 \frac{S(S+1)}{3} \left( \frac{l^2}{8\pi D_1 \tau} \right)^{1/2}, \quad (11)$$

where, for a one-dimensional system,

$$F_0(\theta) = 3 \frac{g\beta}{l^3} \xi(3) (1 - 3 \cos^2 \theta),$$

$$F_1(\theta) = 6 \frac{g\beta}{l^3} \xi(3) \sin \theta \cos \theta,$$

$$F_2(\theta) = 3 \frac{g\beta}{l^3} \xi(3) \sin^2 \theta. \quad (12)$$

At  $\theta_c$  we have  $F_0(\theta) = 0$ , and the ESR linewidth, which is determined by the nonsecular terms  $\Psi_{dm}$ , becomes

$$\Delta H_m = \frac{2}{\gamma} \int_0^\infty (\Psi_{d1} + \Psi_{d2}) d\tau$$

$$= \frac{g^3 \beta^3}{\hbar} \frac{3S(S+1) \xi^2(3)}{l^6} \left( \frac{l}{8D_1 \omega_0} \right)^{1/2}$$

$$\times (10\sqrt{2} \sin^2 \theta \cos^2 \theta + \sin^4 \theta). \quad (13)$$

On the substitution  $\theta = 54.7^\circ$ , this expression agrees numerically with (8), or, in the notation of Ref. 13, it becomes

$$\Delta H_m = 1,268 \Omega_1^2 (l^2/D_1 \omega_0)^{1/2}. \quad (14)$$

For a system whose Hamiltonian also includes a single-ion anisotropy of the form

$$\mathcal{H}_i = A \sum_j S_{zj}^2,$$

there can be additional contributions to  $\Psi^s$  and  $\Psi_{dm}$ . In this case

$$\Delta H_0 = \Delta H_s + \Delta H_{si}$$

$$= \frac{1}{\gamma} \left( \frac{\pi}{2} \right)^{1/2} \frac{1}{\omega_e} \left\{ M_2 + \frac{1}{20} \frac{A^2}{\hbar^2} [4S(S+1) - 3] \right\}, \quad (15)$$

and expression (14) becomes<sup>13</sup>

$$\Delta H_m = 1,268 \left\{ \Omega_1^2 + \frac{9}{20} \frac{g^2 \beta^2}{l^3} \xi(3) [4S(S+1) - 3] \frac{A}{\hbar^2} \right.$$

$$\left. + \frac{1}{20} \frac{A^2}{\hbar^2} [4S(S+1) - 3] \right\} \left( \frac{l^2}{D_1 \omega_0} \right)^{1/2}. \quad (16)$$

Thus, depending on the sign of the parameter  $A$  of the angular coefficient in expression (16), the presence of single-ion anisotropy can lead to a value of  $\Delta H_m = K_1 \nu_0^{-1/2}$  that can be larger or smaller than the value for  $A = 0$ .

To determine the values of  $J$  and  $A$  in TMAMC with the aid of expressions (15) and (16), we used the experimental value of the angular coefficient (Fig. 1)  $K = 1080$  Oe·GHz<sup>1/2</sup> and the value  $\Delta H_0 = 150$  Oe. These values should be multiplied by 3<sup>1/2</sup>, since the linewidth was measured from recordings of the first derivative of the absorption line. The relation between the diffusion coefficient  $D$  and  $J$  was taken in the form  $D_1 = 1.33[S(S+1)]^{1/2} J^2 k / \hbar$ , as has been obtained in computer calculations for one-dimensional systems.<sup>14</sup>

The observed frequency dependence of the ESR linewidth in TMAMC is described by parameter values  $J = 0.36$  K and  $A = -0.188$  K, with  $l = b/2$ . This value of  $J$  is in good agreement with the values obtained from calculations of the angular dependence of the ESR linewidth of this crystal at a frequency of  $\sim 10$  GHz and from specific heat studies.<sup>10</sup> The value of  $A$  is somewhat larger than the values given for this parameter in Refs. 10 and 16, but it does agree in sign.

#### b) CMC

In CMC and ESR linewidth proportional to  $\nu^{-1/2}$  is observed only in the high frequency part of the investigated frequency range. The slope of the linear part of the curve (Fig. 2) and the cutoff are 440 Oe·GHz<sup>1/2</sup> and 35 Oe, respectively. The calculated values of these parameters from expressions (15) and (16) with  $J = 3$  K,  $A = 0.061$  cm<sup>-1</sup> (Ref. 16), and  $l = a/2 = 4.55 A$  are 445 Oe·GHz<sup>1/2</sup> and 31 Oe. The nondiffusional contribution to  $\Delta H$  agrees with the results of Ref. 6, in which a detailed study was made of the angular dependence of the linewidth in CMC.

The deviation from the straight-line behavior  $\Delta H = K_1 \nu^{-1/2}$  in the low frequency part of the investigated range in Fig. 2 could be due to a cutoff (5) of the intrachain spin diffusion. Possible mechanisms for such a cutoff have been analyzed previously for cases including CMC. In one-dimensional systems they usually involve the intrachain dipole-dipole interaction, in which case the characteristic time is  $\tau_2 = (\gamma \Delta H)^{-1} \sim 10^{-10}$  s. Analysis of the shape of the resonance absorption line<sup>2</sup> in CMC also yields a value  $\tau_2 = 10^{-10}$  s, here due to the presence of the weak intrachain exchange interaction  $J'$ . Effective suppression of the intrachain diffusion can also occur when a rapidly relaxing impurity is present in the chain.

When the resonance linewidth is described in the above formalism with allowance for cutoff (5), we get the following expression for its frequency dependence, which is governed by the nonsecular dipole-dipole terms:

$$\Delta H_{1/2} = \frac{2}{\gamma} \int_0^{\infty} [(\Psi_{d_1} + \Psi_{d_2})] d\tau$$

$$= \tau_2^{1/2} \left[ C_1 \left( \frac{1 + (1 + (\omega_0 \tau_2)^2)^{1/2}}{1 + (\omega_0 \tau_2)^2} \right)^{1/2} \right]$$

$$+ C_2 \left( \frac{1 + (1 + (2\omega_0 \tau_2)^2)^{1/2}}{1 + (2\omega_0 \tau_2)^2} \right)^{1/2} \quad (17)$$

where

$$C_{1,2} = [2F_{1,2}(\theta)]^2 \frac{S(S+1)}{3} \left( \frac{l^2}{8\pi D_1} \right)^{1/2}, \quad l = \frac{a}{2} 4.55 \text{ \AA}.$$

A good description of the experimental results for the minimum linewidth in the  $ab$  plane of the crystals (Fig. 2) is obtained with a value  $\tau_2 = 5 \cdot 10^{-12}$  s considerably shorter than the time determined in Ref. 2. For comparison, we also show in Fig. 2 the calculated frequency dependence of the linewidth for the slightly different values  $\tau_2 = 2.5 \cdot 10^{-12}$  and  $7.5 \cdot 10^{-12}$ , demonstrating the sharp dependence of the frequency dependence on this parameter, especially in the low-frequency region.

A possible explanation of why  $\tau_2$  is so short might be the presence of an appreciable dipole-dipole interaction between chains in CMC on account of the structural features of this compound. As we have mentioned, the degree of spatial isolation of the chains, defined as the ratio of the interchain to intrachain distances, is 1.3 in CMC, while for TMAMC it is 2.2 and for the classic one-dimensional antiferromagnet (CH<sub>3</sub>)<sub>4</sub>NMnCl<sub>3</sub> (TMMC) it reaches 2.8. An estimate of the isotropic part of the dipole-dipole interaction in CMC shows that when only 10 coordination spheres are taken into account, the field produced at the Mn<sup>2+</sup> ion in the chain by the ions of the neighboring chains is almost twice as high as the intrachain field. It is evidently this interchain dipole-dipole interaction in CMC that cuts off the diffusion regime in the dissipation of spin correlations in the chain.

#### c) BaMnF<sub>4</sub>

In two-dimensional magnetic systems the long-time behavior  $\Psi(\tau) \sim \tau^{-1}$  of the correlation function leads to a logarithmic frequency dependence of the resonance linewidth at the "magic" angle. According to the results of Richards and Salamon,<sup>3</sup> who evaluated the parameters  $F_m(\theta)$ , the angular dependence of the linewidth at half-height in the high-temperature limit for a plane square lattice should be of the form

$$\gamma \Delta H_{1/2} = \sqrt{\frac{\pi}{2}} \frac{M_2}{\omega_c} (2 + \sin^2 \theta) + \beta' \ln \frac{\omega_c}{\omega_c} (3 \cos^2 \theta - 1)^2$$

$$+ \beta' \left( \ln \frac{\omega_c}{\omega_0} - 0.577 \right) (10 \sin^2 \theta \cos^2 \theta + \sin^4 \theta). \quad (18)$$

Here the first term, as in (10), takes into account the short-time contribution to the linewidth for  $\tau < \tau_1$ . The second term is the secular diffusion contribution, and  $\omega_c = 1/\tau_2 = \gamma \Delta H$  is the cutoff frequency. The third term describes the nonsecular diffusion contribution for  $m = 1, 2$ . The second moment is

$$M_2 = {}^3/2 S(S+1) g^4 \beta^4 w / \hbar^2,$$

the coefficient  $\beta'$  is

$$\beta' = {}^3/2 S(S+1) (g^4 \beta^4 / \hbar^2) v^2 / 32\pi D_2,$$

and the exchange frequency is<sup>3</sup>

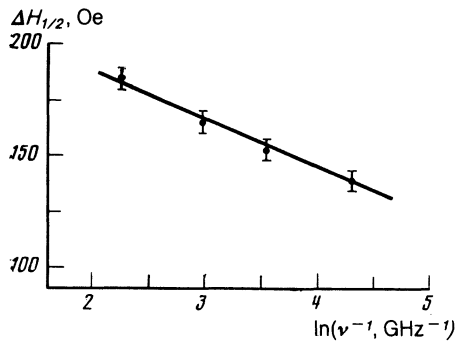


FIG. 3. Frequency dependence of the ESR linewidth  $\Delta H_{1/2}$  at  $\theta_c$  in the  $ab$  plane of the  $\text{BaMnF}_4$  crystal at  $T = 300$  K.

$$\omega_c = \left( 51.2 \frac{S(S+1)}{3} \right)^{1/2} J \frac{k}{\hbar};$$

$$w = \sum_j \left( \frac{1}{r_{ij}} \right)^6;$$

$$v = \sum_j \left( \frac{1}{r_{ij}} \right)^3.$$

On substitution of the numerical values into the third term in (18), which describes the frequency dependence of the linewidth at the "magic" angle, we get a value of the angular coefficient

$$\Delta H = -40.74 J^{-1} \ln \omega_c. \quad (19)$$

Comparison with the experimental value of the slope of  $\Delta H = K_2 \ln \nu_0$  in Fig. 3 gives the value  $J = 5.05$  K. This agrees with the value  $J = 5$  K that was used in interpreting the results on the magnetic susceptibility<sup>17</sup> and antiferromagnetic resonance<sup>18</sup> of this crystal.

On the whole, the angular dependence of the ESR linewidth is also extremely sensitive to the value of  $J$ , on which all three terms in (18) depend. Therefore, we calcu-

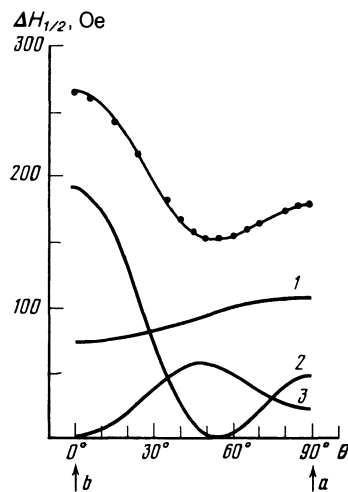


FIG. 4. Angular dependence of the ESR linewidth  $\Delta H_{1/2}$  in  $\text{BaMnF}_4$  at  $T = 300$  K at a frequency of 34.8 GHz. The curve through the points was calculated by Eq. (18); curves 1, 2 and 3 are the respective terms of Eq. (18) for  $J = 5.8$  K.

lated the short-time and diffusion contributions to the linewidth according to (18) for the purpose of finding the value of  $J$  that best describes the angular dependence, which was determined for the  $ab$  plane in our previous paper and found to be described by the empirical formula  $\Delta H = 156 + 27(3 \cos^2 \theta - 1)^2$ .

Figure 4 shows the result of such a calculation for the value  $J = 4.8$  K, which gives extremely good agreement with experiment. The dominant contribution to the linewidth, and that which is responsible for the minimum near  $\theta_c$ , is of course the secular part of the diffusion contribution (curve 2). The short-time contribution, which depends relatively weakly on the angle, comprises about half the linewidth for  $\theta > 40^\circ$ . The nonsecular diffusion contribution (curve 3) comprises only  $\sim 30\%$  of the linewidth in the region near  $\theta_c$ . Therefore, in the case of  $\text{BaMnF}_4$ , it is less reliable to determine  $J$  from the frequency dependence of the linewidth than from the frequency dependence as a whole.

## 5. CONCLUSIONS

Our studies of the frequency dependence of the resonance linewidth at the "magic" angle in low-dimensional magnets clearly confirm that the high-temperature spin dynamics in such systems is of a diffusive nature. The parameter of this frequency dependence, i.e., the angular coefficient obtained with a suitable choice of coordinates and the frequency-independent contribution, can be used to determine the main characteristics of the magnetic system, viz., the diffusion coefficient and the single-ion anisotropy energy. If the relation between  $D$  and  $J$  is single-valued, as it is in a one-dimensional system, one can reliably determine the exchange interaction parameter from these experiments. The angular dependence of the linewidth can also be used to determine the exchange parameter; in combination with the frequency dependence it gives a consistent description of the high-temperature spin dynamics of low-dimensional magnets.

In CMC the interchain dipole-dipole interaction gives a comparatively short existence time for the intrachain correlations,  $\tau_2 = 5 \cdot 10^{-2}$  sec. This interaction is evidently the main source of disruption of the intrachain diffusion, in contrast to the situation in the classic one-dimensional systems of the TMMC type, in which the cutoff is due to the intrachain dipole-dipole and interchain exchange interactions.

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