Two-magnon absorption and characteristics of the spin-wave spectrum of multisublattice antiferromagnets

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The characteristics of the spin-wave spectra and of two-magnon absorption in multisublattice antiferromagnets have been analyzed systematically. Two-magnon absorption by exchange magnons in $CuCl_2 \cdot 2H_2O$ was observed experimentally. A study was made of a new type of dynamic critical point of the spectrum where the group velocity vanishes. These points are due to the interaction of spin waves with one another. It was found that the magnitude of this interaction governs the additional singularities in the density of states which then appear and some components of the effective mass tensor.

INTRODUCTION

A wide range of properties of magnetic materials is governed by the structure of the spectrum and by the characteristics of the density of states of spin waves associated with the presence in their spectrum of critical points where the spin wave group velocity vanishes. Among these critical points we can identify symmetric critical points governed by the symmetry crystal. These critical points can be found without invoking model approximations,[1] which is particularly important in the case of multisublattice magnetic materials. Additional critical points appear at certain values of the intrasublattice and intersublattice exchange constants, and of the magnetic field intensity,[2] in the region where the acoustic and spin wave branches cross,[3] and also when spin wave branches cross one another.[4] The presence of such critical points is in no way related to the crystal symmetry, so that they can be called dynamic.[5]

In experimental studies of spin-wave spectra an important auxiliary parameter is the external static magnetic field, which causes considerable modification of the spectrum. However, the dispersion of all the branches of the spin-wave spectra of multisublattice magnetic materials have been investigated so far only in zero field by the method of inelastic neutron scattering.[6–8] A special feature of multisublattice magnetic materials is the presence in their spin-wave spectra of exchange branches with activation energies which, in the absence of an external field, are governed by the intersublattice exchange interactions and remain finite in the exchange approximation. We recall that the activation energies of acoustic branches are governed by the anisotropy and vanish in the exchange approximation.

One of the effects in which the dispersion dependences of the spin-wave spectrum are manifested is two-magnon absorption, which has no threshold, in contrast to parametric excitation.[9] The intensity of the two-magnon absorption is determined by the number of pairs of magnons which have vectors of the same magnitude, but oppositely directed when the sum of the magnon frequencies has a fixed value equal to the frequency of the incident radiation. Therefore, we can expect an absorption maximum in those parts of the spectrum where the dependence $\omega(\mathbf{k}, H)$ becomes flatter, either because of a modification of the spin-wave spectrum in a magnetic field or because of the quasi-one-dimensional or quasi-two-dimensional nature of the magnetic material. The latter case has been investigated sufficiently thoroughly in the case of two-sublattice antiferromagnets.[10,11]

No experimental or theoretical studies have yet been made of the critical points in the spin-wave spectra of multisublattice antiferromagnets or of the characteristics of the structure of such spectra in magnetic fields or of two-magnon absorption, particularly that due to exchange branches.

These critical points were the subject of a study reported below. We selected a four-sublattice antiferromagnet $CuCl_2 \cdot 2H_2O$ for which the frequency and field dependence of the spectrum of the homogeneous magnetic resonance of the exchange modes had been investigated quite thoroughly.[12]

Our investigation of the magnetic-field dependence of the spin-wave frequencies $\omega(\mathbf{k}, H)$ of a multisublattice antiferromagnet established that the field drastically changes not only the acoustic but also the exchange branches of the spectrum. An indirect confirmation of the flattening of the dispersion curves of the exchange branches was provided by two-magnon absorption by exchange spin waves observed for the first time. The following result, important for further studies of exchange magnons by two-magnon absorption, was obtained: it was found that there were ranges of fields where the absorption intensity was independent of the small parameter DJ^{-1} (D describes the Dzyaloshinskii-Moriya interaction and J is the exchange), because an antiferromagnetic resonance the intensity of the exchange magnon lines were determined by this parameter.[12]

In the case of homogeneous precession of the magnetization a strong interaction of the exchange and acoustic modes of the same symmetry[12] was observed also for spin waves. The range of wave vectors in which this interaction was manifested depended on the magnetic field.

Our results demonstrated the existence of a new type of dynamic critical point in the spin-wave spectrum due to the interaction of spin waves. These points appear when two conditions are satisfied. In the absence of the interaction, they should firstly intersect at a given point in k space and, secondly, at this point the group velocities should be directed along the wave vector $\mathbf{k}$ and antiparallel to one another. An allowance for the interaction (in our specific case this is the Dzyaloshinskii-Moriya interaction) pushes the branches of spin waves apart and gives rise to dynamic criti-

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cal points. Some of the components of the effective mass tensor and the corrections to the density of states associated with such critical points then depend on this interaction.

1. SPIN-WAVE SPECTRUM OF A FOUR-SUBLATTICE ANTIFERROMAGNET

The general form of the Hamiltonian of a magnetic crystal subjected to an external magnetic field is as follows:

$$\mathscr{H}=\sum_{\substack{n,m\\ \alpha,\beta,i,j}} K_{\alpha\beta}^{ij}(n,m)S_{n\alpha}^{i}S_{m\beta}^{j}-\mu_B\sum_{n,\alpha,i,j}H_i g_{ij}^{(\alpha)}S_{n\alpha}^{j}. \qquad (1)$$

In this expression $S_{n\alpha}^{i}$ is the ith component of the operator of the spin in the αth magnetic sublattice and the nth unit cell; $g_{ij}^{(\alpha)}$ is the tensor of the Landé factors of spins in the αth sublattice; μ_B is the Bohr magneton; and the quantities $K_{\alpha\beta}^{ij}$ (n, m) describe the isotropic and antisymmetric exchange interactions as well as the relativistic interactions.

Using the Fourier representation in Eq. (1) and introducing linear combinations of the operators of the ion spins.[1)]

$$\begin{aligned}
\mathbf{F}(\mathbf{k})&=\mathbf{S}_1(\mathbf{k})+\mathbf{S}_2(\mathbf{k})+\mathbf{S}_3(\mathbf{k})+\mathbf{S}_4(\mathbf{k}),\\
\mathbf{L}_1(\mathbf{k})&=\mathbf{S}_1(\mathbf{k})+\mathbf{S}_2(\mathbf{k})-\mathbf{S}_3(\mathbf{k})-\mathbf{S}_4(\mathbf{k}),\\
\mathbf{L}_2(\mathbf{k})&=\mathbf{S}_1(\mathbf{k})-\mathbf{S}_2(\mathbf{k})+\mathbf{S}_3(\mathbf{k})-\mathbf{S}_4(\mathbf{k}),\\
\mathbf{L}_3(\mathbf{k})&=\mathbf{S}_1(\mathbf{k})-\mathbf{S}_2(\mathbf{k})-\mathbf{S}_3(\mathbf{k})+\mathbf{S}_4(\mathbf{k}),
\end{aligned} \qquad (2)$$

we can represent the Hamiltonian of Eq. (1) in the form

$$\mathscr{H}=\sum_{\mathbf{k}}\{\mathscr{H}_0(\mathbf{k})+\delta\mathscr{H}(\mathbf{k})\}. \qquad (3)$$

Here, $\mathscr{H}_0(\mathbf{k})$ also contains invariants of the components of the operators $F(\mathbf{k})$ and $L_\alpha(\mathbf{k})$ similar to those in the $k=0$ case[12]:

$$\begin{aligned}
\mathscr{H}_0(\mathbf{k})=&\sum_i\Big\{J_{0i}(\mathbf{k})F_i(\mathbf{k})F_i(-\mathbf{k})\\
&+\sum_{\sigma=1,2,3}J_{\sigma i}(\mathbf{k})L_{\sigma i}(-\mathbf{k})L_{\sigma i}(\mathbf{k})\Big\}\\
&+D_{01}(\mathbf{k})F_x(-\mathbf{k})L_{2z}(\mathbf{k})+D_{02}(\mathbf{k})F_z(-\mathbf{k})L_{2x}(\mathbf{k})\\
&+D_{03}(\mathbf{k})L_{1x}(-\mathbf{k})L_{3z}(\mathbf{k})+D_{04}(\mathbf{k})L_{1z}(-\mathbf{k})L_{3x}(\mathbf{k})\\
&-N^{1/2}\Delta(\mathbf{k})\{g_1H_x(F_x(\mathbf{k})\\
&+\tau_1L_{2z}(\mathbf{k}))+g_2H_yF_y(\mathbf{k})+g_3H_z(F_z(\mathbf{k})+\tau_3L_{2x}(\mathbf{k}))\}.
\end{aligned} \qquad (4)$$

In the above equation the following notation is used: $g_i=\mu_B g_{ii}$, $\tau_1=g_{xz}g_{xx}^{-1}$, $\tau_3=g_{zx}g_{zz}^{-1}$. The relationship between the quantities $J(\mathbf{k})$ and $D(\mathbf{k})$ and the constants $K_{\alpha\beta}^{ij}(\mathbf{k})$ is readily established with the aid of Eq. (2). The parameters $D(\mathbf{k})$ describing the Dzyaloshinskii interaction include contributions of the antisymmetric parts of $K_{\alpha\beta}^{ij}(\mathbf{k})$ (antisymmetric exchange) as well as of the symmetric parts, which are of purely relativistic origin.[13] In the case of some antiferromagnets the contribution of the latter may predominate.

The nature of $\delta\mathscr{H}(\mathbf{k})$ depends on the direction of the vector **k** and in each specific case a separate symmetry analysis is needed. In particular, at low values of **k** we can easily show that the terms occurring in $\delta\mathscr{H}(\mathbf{k})$ are associated with the inhomogeneous Dzyaloshinskii interaction and with the purely relativistic interactions. It should be noted that calculations of the spin-wave spectrum of yttrium iron garnet based on the Hamiltonian expressed in terms of irreducible combinations of spins (such as in the $\mathbf{k}=0$ case) agree well with a rigorous numerical calculation of the magnon spectrum using the complete Hamiltonian of a 20-sublattice ferromagnet.[14] Bearing this point in mind we shall ignore the terms occurring in $\delta\mathscr{H}(\mathbf{k})$. A rigorous symmetry approach to determination of the structure of the Hamiltonian, utilizing the basis functions of the irreducible representation of the group of the wave vector **k**, can be found in Ref. 15.

In the absence of a field in $CuCl_2\cdot 2H_2O$ we can expect nonzero values of the main antiferromagnetic vector $\mathbf{L}_i$ oriented along the **a** axis (in our coordinate system the x, y, and z axes correspond to the **a**, **b**, and **c** axes of the crystal system) and of the auxiliary antiferromagnetic vector L_3 oriented along the z axis and due to bending of the sublattice magnetizations in the x-z plane due to the Dzyaloshinskii interaction ($\bar{L}_{3z}\propto DJ^{-1}\bar{L}_{ix}$). Since $\bar{L}_{3z}$ and $\bar{L}_{ix}$ transform in accordance with the irreducible representation Γ_6 of the symmetry group D_{2h}^{7} of the paramagnetic phase of the crystal, it follows that the symmetry of the magnetically ordered phase is Γ_6.[2)] In a magnetic field $\mathbf{H}\|\mathbf{x}$ (irreducible representation Γ_2) there are equilibrium values of $\bar{F}_x$ and $\bar{L}_{2z}$ which transform in accordance with the same irreducible representation so that a collinear magnetically ordered phase Γ_{26} is obtained.

When the field is $H_{sf}\sim(g\mu_B)^{-1}(JA)^{1-2}$, a spin-reorientation phase transition takes place and the vector $\mathbf{L}_i$ becomes parallel to the y axis, which corresponds to a magnetically ordered Γ_{27} phase or a spin-flop phase. In a field of the order of the exchange field a second-order phase transition takes place, which involves collapse of the magnetic sublattices (spin-flip transition) when $\mathbf{L}_i$ vanishes and a magnetically ordered Γ_2 phase or a flip phase is formed. The magnetic cell of this phase in $CuCl_2\cdot 2H_2O$ is identical with the crystallographic unit cell and the number of sublattices is halved, so that instead of four magnetic resonance modes there are only two.

In the case of antiferromagnets, which include $CuCl_2\cdot 2H_2O$, the quantities J and D are related by $J\gg D\gg A\sim D^2J^{-1}$, where A is a purely relativistic anisotropic interaction. Hence, it follows that the auxiliary antiferromagnetic vectors $\mathbf{L}_2$ and $\mathbf{L}_3$ are small compared with S, which is the sublattice spin throughout the range of existence of magnetically ordered phases. In the problems of interest to us an allowance for such fine details of the magnetic structure is unimportant. Therefore, we shall carry out specific calculations employing a rough description of the magnetic order on the assumption that in the case of the Γ_6 and Γ_{26} phases we have $\bar{L}_{ix}/4S=1$, whereas in the case of the Γ_{27} phase, we find that

$$\bar{L}_{1y}/4S=\delta=(1-\beta^2)^{1/2},\ \bar{F}_x/4S=\beta=HH_{AF}^{-1},$$

where H_{AF} is the field of the antiferromagnetic exchange between the sublattices 1 and 3. In the case of the Γ_2 phase, we have $\bar{F}_x/2S=1$. The form of a magnetic unit cell is shown in Ref. 12. Experimental and theoretical investigations of the frequency and field dependence of homogeneous oscillations of exchange and acoustic modes, reported in Ref. 12, have made it possible to determine the ferromagnetic exchange field H_F between ions 1 and 2, and the value of the parameter D.

We shall calculate the spin-wave spectrum using the method of second quantization developed for multisublattice magnetic materials in Ref. 16. Specific calculations will be made in the nearest-neighbor approximation allowing only for the interactions between pairs of ions 12 and 13. We shall not discuss details of the calculations, which were carried out in Ref. 4, allowing fully for the magnetic structure, but simply give the frequencies of spin waves for different magnetically ordered phases.

Γ_{26} phase (collinear phase)

The energies of the exchange branches in the spin-wave spectrum are described by the expressions

$$\omega_{1256}^{(\pm)}(\mathbf{k},H)=(E_2(\mathbf{k})E_3(\mathbf{k}))^{1/2}\pm gH. \qquad (5)$$

In view of the above approximations, this expression is valid in the range of fields $H_A \lesssim H \lesssim H_{sf}$, where H_A is the characteristic anisotropy field. We shall allow for the anisotropy fields only in those cases when they should give a nonzero acoustic mode activation energy:

$$\omega_{3478}^{(\pm)}(\mathbf{k},H)=2^{-1/2}\{q_2p_2+q_3p_3+2p_{23}{}^2\pm[(q_2p_2+q_3p_3+2p_{23}{}^2)^2 -4(q_2q_3-p_{23}{}^2)(p_2p_3-p_{23}{}^2)]^{1/2}\}^{1/2}, \qquad (6)$$

where

$$q_2=E_1(\mathbf{k})+gH_{Ay},\quad p_3=E_1(\mathbf{k})+gH_{Az},$$
$$p_{23}=gH,\quad p_2=q_3=E_0(\mathbf{k}).$$

Combinations of the exchange integrals

$$E_{0i}(\mathbf{k})=8S(J_{0i}(\mathbf{k})-J_{1x}(0))$$

obtained using the above approximations are of the form

$$E_0(\mathbf{k})=\frac{1}{2}g\{H_{AF}(1+\gamma_{13}(\mathbf{k}))+H_F(1-\gamma_{12}(\mathbf{k}))\},$$
$$E_1(\mathbf{k})=\frac{1}{2}g\{H_{AF}(1-\gamma_{13}(\mathbf{k}))+H_F(1-\gamma_{12}(\mathbf{k}))\},$$
$$E_2(\mathbf{k})=\frac{1}{2}g\{H_{AF}(1+\gamma_{13}(\mathbf{k}))+H_F(1+\gamma_{12}(\mathbf{k}))\}, \qquad (7)$$
$$E_3(\mathbf{k})=\frac{1}{2}g\{H_{AF}(1-\gamma_{13}(\mathbf{k}))+H_F(1+\gamma_{12}(\mathbf{k}))\}.$$

The structure factors $\gamma(\mathbf{k})$ are described by the expressions

$$\gamma_{12}(\mathbf{k})=\cos\frac{\mathbf{ka}}{2}\cos\frac{\mathbf{kb}}{2},\qquad \gamma_{13}(\mathbf{k})=\cos\frac{\mathbf{kc}}{2}, \qquad (8)$$

where **a**, **b**, and **c** are the constants of a unit magnetic cell and $g\equiv g_1=\mu_B g_{xx}$.

In the expressions for $D(\mathbf{k})$ deduced from Eq. (4) we need retain only the contribution of the antisymmetric exchange of pairs[12]; we then obtain

$$D_1(\mathbf{k})=-D_2(\mathbf{k})=D_3(\mathbf{k})=-D_4(\mathbf{k})=gD\gamma_{12}(\mathbf{k}). \qquad (9)$$

The values of all the parameters we used are[12,17,18]

$$H_{AF}=15\,\mathrm{T},\ H_F=3.65\,\mathrm{T},\ D=0.25\,\mathrm{T},\ g_{xx}=2.187,$$
$$H_{Ay}=g^{-1}(E_{1y}+D_3{}^2/E_{3z})=0.028\,\mathrm{T},\ H_{Az}=g^{-1}E_{1z}=0.086\,\mathrm{T}. \qquad (10)$$

It should be noted that in the case of acoustic branches of the spin-wave spectrum the nonlinear field dependence exhibited by the collinear phase is important only in the range $ka \ll 1$. At high values of **k** the energies $\omega_{3478}^{(\pm)}(\mathbf{k},H)$ vary linearly with the field. Figure 1 shows how the frequencies of all spin waves depend on **k** in a field $H = 0.4$ T, plotted for the wave vectors $\mathbf{k}\|\mathbf{a}$ and $\mathbf{k}\|\mathbf{c}$.

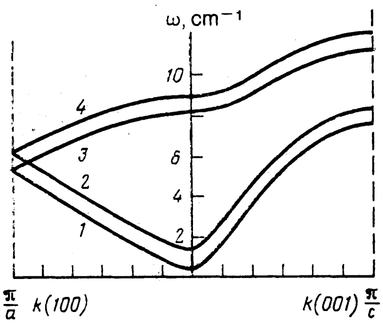

FIG. 1. Γ_{26} phase, $H = 4$ kOe: 1) $\omega_{3478}^{(-)}$, 2) $\omega_{3478}^{(+)}$, 3) $\omega_{1256}^{(-)}$, 4) $\omega_{1256}^{(+)}$.

Γ_{27} phase (spin-flop phase)

In this phase both acoustic and exchange branches have the same symmetry in pairs. The Dzyaloshinskii interaction plays the role of the coupling parameters of these modes. The spin-wave energy is

$$\omega_{3456}^{(\pm)}(\mathbf{k},H)=2^{-1/2}\{q_1p_1+q_2p_2-2T^2\pm[(q_1p_1-q_2p_2)^2 +4T^2E_2{}^2(0)]^{1/2}\}^{1/2},$$

where

$$p_1=E_2(\mathbf{k})\delta^2+\beta^2E_3(\mathbf{k}),\ p_2=E_1(\mathbf{k})\delta^2+\beta^2E_0(\mathbf{k}),$$
$$q_1=E_3(\mathbf{k}),\ q_2=E_0(\mathbf{k}), \qquad (11)$$
$$T=g\delta D[\gamma_{12}(\mathbf{k})-\beta^2H_{AF}(H_{AF}+H_F)^{-1}\gamma_{13}(\mathbf{k})].$$

The expression for the spin-wave energy $\omega_{1278}^{(\pm)}(\mathbf{k},H)$ can be obtained from Eq. (11) by modification of the indices $1\to 0$ and $2\to 3$ of the quantities p and q. We then have

$$p_0=\delta^2E_3(\mathbf{k})+\beta^2E_2(\mathbf{k}),\ p_3=\delta^2E_0(\mathbf{k})+\beta^2E_1(\mathbf{k}),$$
$$q_0=E_2(\mathbf{k}),\ q_3=E_1(\mathbf{k})+g(H_{Az}-H_{Ay}).$$

The characteristic features of the behavior of the spin-wave spectra of the spin-flop phase are demonstrated in Figs. 2–4 for fields of 4, 7, and 10 T.

The Dzyaloshinskii interaction gives rise to additional extrema in the spin-wave dispersion laws within the Brillouin zone. The positions of the extrema depend on the mag-

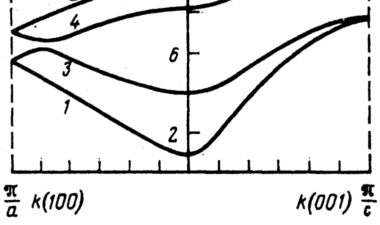

FIG. 2. Γ_{27} phase, $H = 40$ kOe: 1) $\omega_{1278}^{(-)}$, 2) $\omega_{1278}^{(+)}$, 3) $\omega_{3456}^{(-)}$, 4) $\omega_{3456}^{(+)}$.

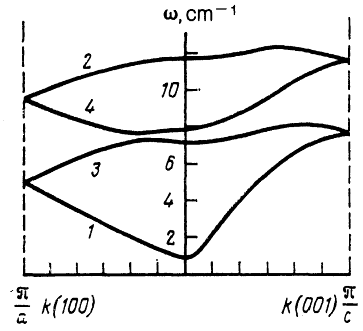

FIG. 3. Γ_{27} phase, $H = 70$ kOe.

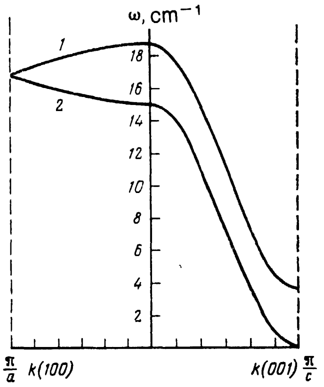

FIG. 5. Γ_2 phase, $H = 150$ kOe: 1) ω_{12}; 2) ω_{34}.

netic field and can shift toward smaller and larger wave vectors. Since lifting of the degeneracy is due to the Dzyaloshinskii interaction, the smallest separation between the frequencies is proportional to D, as in the homogeneous case.[12] Moreover, we can assume that the absence of crossing of the frequencies of acoustic and exchange branches of the same symmetry in a field H_m and $\mathbf{k} = 0$ (Fig. 3 in Ref. 12) represents a special case of lifting of the degeneracy of these frequencies in the $\mathbf{k}$ space in fields higher and lower than H_m. The characteristic features of the density of states in this case will be discussed later.

In the spin-flop phase there is a set of points in the $\mathbf{k}$ space where the spin-wave energy is independent of the magnetic field (in the approximation we have adopted). This is true, in particular, of two planes of the Brillouin zone boundary perpendicular to z.

Γ_2 phase (spin-flip phase)

The Hamiltonian of the magnetic subsystem of a crystal in this phase is obtained from Eq. (4) by assuming that the sublattices 1 and 3 and also 2 and 4 are identical. Then, the constants J_1, J_3, D_3, and D_4 vanish and there are only two types of irreducible operators $\mathbf{F} = \mathbf{S}_1 + \mathbf{S}_2$ and $\mathbf{L}_2 = \mathbf{S}_1 - \mathbf{S}_2$.

The spin-wave frequencies in this phase are given by

$$\omega_{34}(\mathbf{k}, H) = E_0(\mathbf{k}) - E_0(0) + gH,$$
$$\omega_{12}(\mathbf{k}, H) = E_2(\mathbf{k}) - E_0(0) + gH. \qquad (12)$$

The phase transition from the spin-flip to the spin-flop phase is of second order. The soft mode in the flip phase, which vanishes at the phase transition point, is the spin-wave

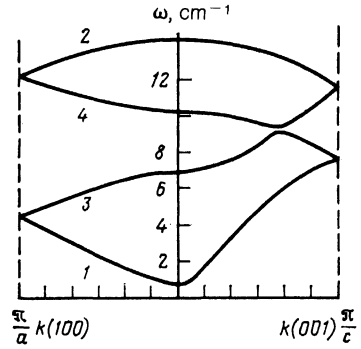

FIG. 4. Γ_{27} phase, $H = 100$ kOe.

frequency $\omega_{34}(k_{zB}, H)$ with the wave vector $\mathbf{k}\|\mathbf{z}$ lying at the boundary of the Brillouin zone of the flip phase $k_{zB} = \pi c_0^{-1}$, where c_0 is the constant of the crystallographic unit cell. Figure 5 shows how the frequencies of the spin waves Γ_{34} and Γ_{12} depend on $\mathbf{k}$ in the flip-transition field $H_f = H_{AF}$.

2. CRITICAL POINTS OF SPIN-WAVE SPECTRA OF MULTISUBLATTICE MAGNETIC MATERIALS AND SINGULARITIES OF THE DENSITY OF STATES

In discussing the specific features of the density of states typical of multisublattice magnetic materials it is necessary to distinguish two sets of critical points. Firstly, there are dynamic critical points, the appearance of which is due to the interaction of spin waves with one another (obviously, such critical points can exist in the case of multisublattice magnetic materials, which are characterized by a large number of branches of spin waves). Secondly, there are symmetric critical points that can be found without invoking model approximations, which is particularly important in the case of multisublattice magnetic materials.

We shall begin our analysis from the symmetric critical points. The criterion of existence of zero-slope points in the spectra of quasiparticles is formulated in Ref. 1 for the case of magnetic symmetry groups. Table I gives the results of an analysis, based on the approach of Ref. 1, of the components of the spin-wave group velocity of vector $\mathbf{v} = \partial\omega/\partial\mathbf{k}$ at high-symmetry points of the Brillouin zone of our crystal, obtained for all possible magnetically ordered phases with the given orientation of the magnetic field.[19] (The plus sign corresponds to a nonzero component of the group velocity, whereas the minus sign corresponds to a zero component; $\mathbf{b}_1$, $\mathbf{b}_2$, and $\mathbf{b}_3$ are the reciprocal lattice vectors and the parameters μ and ν vary within the limits $(0 < \mu, \nu < 1/2.)$

It is clear from Table I that in the absence of a magnetic field the symmetric critical points (in the Γ_6 phase) are two points in the Brillouin zone: $\mathbf{k}_0 = 0$ and $\mathbf{k}_0 = (\mathbf{b}_1 + \mathbf{b}_2)/2$. Application of a magnetic field $\mathbf{H}\|a\|\mathbf{x}$ does not suppress these critical points of the phases Γ_{26}, Γ_{27}, and Γ_2. The density of states corresponding to these critical points has a trivial singularity of the $|\omega - \omega_0|^{1/2}$ type. The specific nature of the magnetic groups of all four magnetically ordered phases is such that vanishing or nonvanishing of the same components of the group velocity occurs for all the branches of spin waves irrespective of their symmetry at any high-symmetry points in the Brillouin zone.

TABLE I. Components of group velocities of spin waves at high-symmetry points of Brillouin zone of different magnetically ordered phases.

$\mathbf{k}_0$	Γ_6			$\Gamma_{26}\Gamma_2$			Γ_{27}		
	v_x	v_y	v_z	v_x	v_y	v_z	v_x	v_y	v_z
$\mathbf{k}=\mu\mathbf{b}_1$	+	−	−	+	−	−	+	−	−
$\mathbf{k}=\mu\mathbf{b}_1+\mathbf{b}_2/2$	+	+	+	+	+	+	+	−	−
$\mathbf{k}=\mu\mathbf{b}_1+\mathbf{b}_3/2$	+	+	+	+	+	+	+	−	−
$\mathbf{k}=\mu\mathbf{b}_1+(\mathbf{b}_2+\mathbf{b}_3)/2$	+	−	−	+	−	−	+	−	−
$\mathbf{k}=\mu\mathbf{b}_2$	−	+	−	+	+	+	−	+	−
$\mathbf{k}=\mu\mathbf{b}_2+\mathbf{b}_1/2$	+	+	+	+	+	+	−	+	−
$\mathbf{k}=\mu\mathbf{b}_2+\mathbf{b}_3/2$	−	+	−	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_2+(\mathbf{b}_1+\mathbf{b}_3)/2$	+	+	+	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_3$	−	−	+	+	+	+	−	−	+
$\mathbf{k}=\mu\mathbf{b}_3+\mathbf{b}_1/2$	−	−	+	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_3+\mathbf{b}_2/2$	+	+	+	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_3+(\mathbf{b}_1+\mathbf{b}_2)/2$	+	+	+	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_1+\nu\mathbf{b}_2$	+	+	−	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_2+\nu\mathbf{b}_3$	−	+	+	−	+	+	−	+	+
$\mathbf{k}=\mu\mathbf{b}_2+\nu\mathbf{b}_3+\mathbf{b}_1/2$	−	+	+	−	+	+	−	+	+
$\mathbf{k}=\mu\mathbf{b}_1+\nu\mathbf{b}_3$	+	−	+	+	+	+	+	+	+
$\mathbf{k}=\mu\mathbf{b}_1+\nu\mathbf{b}_3+\mathbf{b}_2/2$	+	−	+	+	+	+	+	+	+
$\mathbf{k}=0$	−	−	−	−	−	−	−	−	−
$\mathbf{k}=\mathbf{b}_1/2$	+	−	−	+	−	−	+	+	−
$\mathbf{k}=\mathbf{b}_2/2$	−	+	+	−	+	+	+	+	−
$\mathbf{k}=\mathbf{b}_3/2$	−	+	+	−	+	+	+	−	+
$\mathbf{k}=(\mathbf{b}_1+\mathbf{b}_2)/2$	−	−	−	−	−	−	−	−	−
$\mathbf{k}=(\mathbf{b}_1+\mathbf{b}_3)/2$	−	+	+	+	+	+	+	−	−
$\mathbf{k}=(\mathbf{b}_2+\mathbf{b}_3)/2$	+	+	−	+	+	+	+	−	−
$\mathbf{k}=(\mathbf{b}_1+\mathbf{b}_2+\mathbf{b}_3)/2$	+	−	−	+	−	−	+	−	−

The fact that some derivatives $\partial\omega(\mathbf{k}_0)/\partial\mathbf{k}_0$ differ from zero on transition to points of higher symmetry is due to the fact that degeneracy of the spin-wave energies appears at these points. For example, it is clear from Table I, that in the Γ_{27} phase the derivative in question is $\partial\omega(\mathbf{k}_0)/\partial k_0^y=0$ for $\mathbf{k}_0=\mu\mathbf{b}_1$, whereas at the point $\mathbf{k}_0=\mathbf{b}_1/2$ this derivative is nonzero. A symmetry analysis shows that if $\mathbf{k}_0=\mathbf{b}_1/2$, then the states of spin waves transform in accordance with the two-dimensional irreducible representation and, consequently, the energies corresponding to them are degenerate, whereas if $\mathbf{k}_0=\mu\mathbf{b}_1$, all the states transform in accordance with one-dimensional irreducible representations.

In a multisublattice magnetic material we may encounter a situation in which the branches of the spectrum of magnon modes considered in the exchange approximation cross at some point or region of the **k** space (this is known as the crossover situation). An allowance for the anisotropic interactions sometimes lifts the degeneracy at this point or region and pushes apart the branches of spin waves.[4] In turn, this can give rise to extremal points of the dispersion curves along the direction **k** and cause vanishing of the corresponding component of the group velocity. Vanishing of the other components can follow, for example, from symmetry considerations. The positions of such dynamic critical points, due to the interaction of spin waves, depends on the magnetic field and may shift to the range of small or large wave vectors. Lifting of the degeneracy in the crossover region need not give rise to dynamic critical points which may happen, for example, in some cases of a magnetoacoustic resonance or in the polariton effects. These dynamic critical points appear if the group velocities of two branches have opposite directions before allowance for the interaction.

We shall consider the example of a crossover situation which occurs in the case of exchange and acoustic branches of spin waves $\omega_{3456}^{(\pm)}(\mathbf{k},H)$ of the Γ_{27} (spin-flop) phase of $CuCl_2\cdot 2H_2O$ (Ref. 4). Using symmetry considerations, we can demonstrate rigorously that lifting of the degeneracy in this case is due to the Dzyaloshinskii interaction, i.e., it is due to the off-diagonal components of the Hamiltonian DS_iS_j $(i\neq j)$, whereas the purely relativistic diagonal anisotropic interactions do not push apart the branches.[15]

If we ignore the Dzyaloshinskii interaction, then in the case of the energies $\omega_{3456}^{(+)}(\mathbf{k},H)$ and $\omega_{3456}^{(-)}(\mathbf{k},H)$ we have a crossover situation on some surface in the **k** space found from the condition

$$\gamma_{13}(\mathbf{k})=H_FH_{AF}^{-1}\beta^{-2}\gamma_{12}(\mathbf{k}). \tag{13}$$

From now on we shall distinguish two cases: a) $\beta^2>H_FH_{AF}^{-1}$ or $H>H_m=(H_FH_{AF})^{1/2}$; b) $\beta^2<H_FH_{AF}^{-1}$. An allowance for D pushes apart the branches of spin waves and gives rise to extremal points for each of them along certain directions (see Fig. 4 for case *a* and Fig. 2 for case *b*). We can identify critical points among these extrema by invoking additional considerations (symmetry or model). For example, in case *a*) it follows from symmetry considerations that for the wave vectors $\mathbf{k}=\mu\mathbf{b}_3$ the group velocity components $v_x=\partial\omega_{3456}^{(\pm)}/\partial k^x$ and $v_y=\partial\omega_{3456}^{(\pm)}/\partial k^y$ vanish, whereas the third component of the velocity is $v_z=\partial\omega_{3456}^{(\pm)}/\partial k^z$ because of the Dzyaloshinskii interaction. The remaining points in **k** space, satisfying Eq. (13), are not critical in the case *a*), because in this case the values of v_x and v_y are nonzero. In this sense we are dealing with an isolated critical point for $H>H_m$. As shown below, for $H<H_m$, there are critical points located along a certain line in the **k** space, i.e., there are nonisolated critical points. We shall consider separately the isolated and nonisolated critical points.

A. Isolated critical points $\mathbf{k}_0$ $(0,0,\mathbf{k}_{0z})=\mu\mathbf{b}_3$.

We shall find k_{0z} from

$$\frac{\partial\omega_{3456}^{(\pm)}(\mathbf{k},H)}{\partial k_z}=-\frac{c}{2}\sin\frac{k_zc}{2}\frac{\partial\omega_{3456}^{(\pm)}(\mathbf{k},H)}{\partial\gamma_{13}}=0. \tag{14}$$

Near this critical point we can represent $\omega_{3456}^{(\pm)}$ in the form

$$\omega_{3456}^{(\pm)2}(\mathbf{k},H)=\omega_0^{(\pm)2}+(\gamma_{13}-\bar{\gamma}_{13})^2/m^{(\pm)}+\varkappa_\perp^2/m_\perp^{(\pm)}, \quad (15)$$

where $\varkappa_\perp^2=\varkappa_x^2+\varkappa_y^2$, $\varkappa_i=1/2k_ia_i$, $\omega_0^{(\pm)}=\omega_{3456}^{(\pm)}(k_{0z}H)$, $\bar{\gamma}_{13}=\gamma_{13}(k_{0z})$,

$$(m^{(\pm)})^{-1}=\pm H_{AF}[(\beta^4H_2)^2-(\delta^4H_F)^2]^{3/2}/4\delta\beta^8H_2D,$$
$$(m_\perp^{(\pm)})^{-1}=\pm H_2H_F^2\delta^4/H_{AF}\beta^4. \quad (16)$$

We can readily see from Eq. (16) that $m_{zz}=m^{(\pm)}\omega_0^{(\pm)}$ is a component of the effective mass tensor proportional to the ratio DJ^{-1}, whereas the components m_{xx} and m_{yy} are independent of the parameter D.

The correction to the density of states $\delta g(\omega)$ due to a dynamic critical point, resulting from the interaction between exchange and acoustic branches of spin waves, is described by the expression

$$\delta g(\omega)=\frac{4\omega m_\perp^{(\pm)}}{\pi^2}\{\arccos\bar{\gamma}_{13}^{(\pm)}-\arccos(\bar{\gamma}_{13}^{(\pm)}+\theta(\omega-\omega_0^{(+)})[m^{(+)}(\omega^2-\omega_0^{(+)2})]^{1/2}+\theta(\omega_0^{(-)}-\omega)[m^{(-)}(\omega_0^{(-)2}-\omega^2)]^{1/2})\}. \quad (17)$$

We can readily see that if $\omega\approx\omega_0$, we obtain the usual square-root singularities. Then, the magnitude of the correction $\delta g(\omega)$ is proportional to the small parameter $(DJ^{-1})^{1/2}$.

B. Nonisolated dynamic critical points. If $H<H_m$, Eq. (13) is satisfied for the components k_x and k_y lying along a certain line in the $k_z=$ const plane. We shall be interested in the value $k_z=0$ so that in this case we have $\partial\omega_{3456}^{(\pm)}/\partial k_z=0$. The positions of the dynamic critical points on the $k_z=0$ plane are found from the equation

$$(\partial\omega_{3456}^{(\pm)}/\partial\gamma_{12})_{k_z=0}=0.$$

We shall now consider the most interesting situation when $H\lesssim H_m$. In this case a line of dynamic critical points is close to a circle of radius

$$\varkappa_{0\perp}=(1-\beta^2H_{AF}H_F^{-1})^{1/2}.$$

In this region we can represent $\omega_{3456}^{(\pm)}$ by

$$\omega_{3456}^{(\pm)2}(\mathbf{k},H)=\omega_0^{(\pm)2}+(^1/_2\varkappa_\perp^2-\varkappa_{0\perp}^2)^2/\tilde{m}_\perp^{(\pm)}+\varkappa_z^2/\tilde{m}^{(\pm)}, \quad (18)$$

where

$$(\tilde{m}_\perp^{(\pm)})^{-1}=\pm H_F^3H_2^2/4\delta\beta^2H_{AF}^2D,\ (\tilde{m}^{(\pm)})^{-1}=\tfrac{1}{2}H_{AF}^2\delta^4. \quad (19)$$

It is clear from Eq. (18) that a distinguishing feature of the case under discussion is that near the line of dynamic critical points there is a symmetric critical point $\mathbf{k}=0$. A similar situation in which there are close critical points is considered in Ref. 20. An analysis of the density of states of frequencies of spin waves described by Eq. (19) should be carried out as a function of the relationship between the parameters $\omega_0^{(\pm)}$, $\varkappa_{0\perp}$, $\tilde{m}_\perp^{(\pm)}$, and $\tilde{m}^{(\pm)}$:

1) $\omega_{3456}^{(+)}$, $\omega>\omega_0^{(+)}$, $(\omega^2-\omega_0^{(+)2})\tilde{m}_\perp^{(+)}\leqslant\varkappa_{0\perp}^4$,

$$\delta g^{(+)}(\omega)=4\omega\pi^{-1}(\tilde{m}_\perp^{(+)}\tilde{m}^{(+)})^{1/2}\equiv A^{(+)}\omega;$$

2) $\omega_{3456}^{(+)}$, $\omega>\omega_0^{(+)}$, $(\omega^2-\omega_0^{(+)2})\tilde{m}_\perp^{(+)}>\varkappa_{0\perp}^4$,

$$\delta g^{(+)}(\omega)=\frac{1}{2}A^{(+)}\pi^{-1}\arccos\frac{(\omega^2-\omega_0^{(+)2}-\varkappa_{0\perp}^4/\tilde{m}^{(+)})^{1/2}}{(\omega^2-\omega_0^{(+)2})^{1/2}}; \quad (20)$$

3) $\omega_{3456}^{(-)}$, $\omega<\omega_0^{(-)}$, $(\omega_0^{(-)2}-\omega^2)|\tilde{m}_\perp^{(-)}|<\varkappa_{0\perp}^4$,

$$\delta g^{(-)}(\omega)=\frac{1}{4}A^{(-)}\pi^{-1}\ln\left|\frac{\omega'^2}{\omega_0^{(-)2}-\omega^2}\right|,$$

where ω' is a certain characteristic energy of the order of $\omega_0^{(-)}$;

4) $\omega_{3456}^{(-)}$, $\omega<\omega_0^{(-)}$, $(\omega_0^{(-)2}-\omega^2)|\tilde{m}_\perp^{(-)}|<\varkappa_{0\perp}^4$,

$$\delta g^{(-)}(\omega)=A^{(-)}\pi^{-1}\ln\left|\frac{\omega'[(\varkappa_{0\perp}^4-(\omega_0^{(-)2}-\omega^2)|\tilde{m}_\perp^{(-)}|)^{1/2}+\varkappa_{0\perp}^2]}{|\tilde{m}_\perp^{(-)}\tilde{m}^{(-)}|^{1/2}(\omega_0^{(-)2}-\omega^2)}\right|;$$

5) $\omega_{3456}^{(-)}$, $\omega>\omega_0^{(-)}$,

$$\delta g^{(-)}(\omega)=A^{(-)}\pi^{-1}\ln\left|\frac{\omega'[(\varkappa_{0\perp}^4+(\omega_0^{(-)2}-\omega^2)|\tilde{m}_\perp^{(-)}|)^{1/2}+\varkappa_{0\perp}^2]}{|\tilde{m}_\perp^{(-)}\tilde{m}^{(-)}|^{1/2}(\omega_0^{(-)2}-\omega^2)}\right|.$$

The constant-energy surfaces corresponding to each of these cases are shown in Fig. 6.

There are also dynamic critical points due to a change in the magnetic field, which are present also in two-sublattice magnetic materials.[2] They appear because at some points in **k** space the energy of spin waves is independent of the field

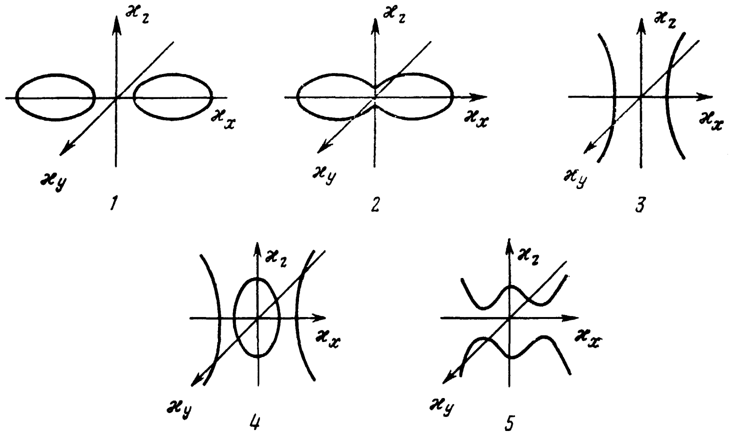

FIG. 6. Sections of constant-energy surfaces plotted for $\omega_{3456}^{(\pm)}(\mathbf{k})$ energies near nonisolated critical points. These are surfaces of revolution about the vertical axis of curves identified as 1–5. The numbers of the curves 1–5 are the same as the numbers of the cases of Eq. (20).

(on the basis of the adopted model). Therefore, a change in the magnetic field on the dispersion curves may create additional extrema associated with flattening of these curves. It should be pointed out that such dynamic critical points appear in the case of acoustic and exchange spin waves. We shall not analyze them in detail, but their appearance in two-magnon absorption due to exchange spin waves will be discussed in the next section.

3. TWO-MAGNON ABSORPTION IN A FOUR-SUBLATTICE ANTIFERROMAGNET: THEORY

The two-magnon absorption in the orthorhombic antiferromagnet which we investigated is of the magnetic-dipole nature. The part of the Hamiltonian describing the interaction of an external alternating magnetic field H_ω with the spin subsystem of interest to us is of the form

$$\Delta\mathscr{H}_{int}=-|h_{i\omega}|\sum_{\nu_1,\nu_2,\mathbf{k}} f_{\nu_1\nu_2}(\mathbf{k})\overset{+}{\xi}_{\nu_1}(-\mathbf{k})\overset{+}{\xi}_{\nu_2}(\mathbf{k}), \qquad (21)$$

where $h_{i\omega}=\mu_B g_{ii} H_{i\omega}$ *and* $f_{\upsilon 1\upsilon 2}(\mathbf{k})$ depend on the polarization of the field H_ω of the ground states of a specific magnetically ordered phase and can be expressed in terms of the coefficients of the u–v Bogolyubov transformation of the corresponding branches of spin waves. In the case of one magnon branch $\nu_1=\nu_2=\nu$ the absorption coefficient is

$$K_\nu(\omega,H)=\frac{\omega|h_\omega|^2\,\mathrm{cth}(\hbar\omega/2T)}{2\pi\hbar c|H_\omega|^2}\int d^3k\,|f_\nu(\mathbf{k})|^2\delta(2\omega_{\mathbf{k}\nu}-\omega). \qquad (22)$$

We shall now analyze the processes of creation of magnon pairs in the case of different polarizations of an external alternating magnetic field, and we shall do this for all the relevant magnetically ordered phases.

Γ_{26} phase

a) $\mathbf{H}_\omega\|\mathbf{x}$. The structure of the Bogolyubov transformation coefficients for this phase is such that the contribution of the exchange branches to the two-magnon absorption vanishes.

b) $\mathbf{H}_\omega\|\mathbf{y}$. In this polarization there are no processes that can create magnon pairs. We shall ignore the possibility of magnon pair creation as a result of decay of homogeneous precession due to three-magnon interactions.

c) $\mathbf{H}_\omega\|\mathbf{z}$. In this polarization the two-magnon absorption coefficient is small and proportional to $(DJ^{-1})^2$.

Γ_{27} phase

a) $\mathbf{H}_\omega\|\mathbf{x}$. In this case the two-magnon absorption will be observed for any two branches of the same symmetry, $\omega_{1278}^{(\pm)}$ and $\omega_{3456}^{(\pm)}$; the process of absorption by the $\omega_{1278}^{(+)}$ branch will be discussed in detail later.

b) $\mathbf{H}_\omega\|\mathbf{y}$. In this polarization we can expect creation of pairs in branches of different symmetry, for example, two exchange branches $\omega_{3456}^{(+)}$ and $\omega_{1278}^{(+)}$.

c) $\mathbf{H}_\omega\|\mathbf{z}$. In this case the absorption coefficient is small, in the same way as in the case of *c*) of the Γ_{26} phase.

Γ_2 phase

a) $\mathbf{H}_\omega\|\mathbf{x}$. Two-magnon absorption should be exhibited by each of the branches ω_{34} and ω_{12}.

b) $\mathbf{H}_\omega\|\mathbf{y}$. There is no two-magnon absorption.

c) $\mathbf{H}_\omega\|\mathbf{z}$. The intensity of two-magnon absorption is low, as in the case of the corresponding cases of collinear and spin-flop phases.

We shall consider in detail the two-magnon absorption in the spin-flop phase due to the exchange mode $\omega_{1278}^{(+)}$, when an external oscillatory field H_ω is oriented along the x axis. In this case, we have

$$f_{1278}(\mathbf{k})\approx\beta\delta H_{AF}\gamma_{13}(\mathbf{k})/\omega_{1278}^{(+)}(\mathbf{k},H).$$

The most important feature is the fact that the amplitude of such two-magnon absorption due to exchange branches does not contain the small factor DJ^{-1}. We shall substitute the above expression in Eq. (22). Bearing in mind $\hbar\omega/2T\sim 10$ and $\cot(\hbar\omega/2T)\sim 1$, we find that simple transformations yield the following expression for the absorption coefficient:

$$K_{1278}^{(+)}(\omega,H)=(8g^2H_{AF}^2\beta^2\delta^4/\pi\hbar c v_0)\,J(\omega,H),$$

where

$$J(\omega,H)=\int_0^1\frac{dx}{(1-x^2)^{1/2}}\int_0^1\frac{dy}{(1-y^2)^{1/2}} \times\int_0^1\frac{z^2\,dz}{(1-z^2)^{1/2}}\,\delta\left\{\omega_{1278}^{(+)2}(x,y,z)-\frac{\omega^2}{4}\right\}.$$

We have made the substitution $\varkappa_i=x_i$ ($i=1, 2, 3$). The function $J(\omega, H)$ is tabulated for different frequencies throughout the full range of fields corresponding to the existence of the spin-flop phase. Figure 7 shows how the two-magnon absorption coefficient depends on the magnetic field, ignoring the attenuation at a fixed frequency of the incident radiation. As expected, the absorption maximum appears at frequencies near twice the energy of spin waves with $k=k_{zB}$ for magnetic fields corresponding to flattening of the spin-wave dispersion law $\omega_{1278}^{(+)}(\mathbf{k},H)$, which is exactly where dynamic critical points associated with the dependence of the spectrum on the magnetic field appear.

We shall consider two-magnon absorption at radiation frequencies ω located near the double energy corresponding to the range of interaction of spin waves. Contributions to this absorption by other parts of the spin-wave spectrum will be ignored. Bearing in mind that $\omega_{3456}^{(+)}(\mathbf{k},H)\approx\omega_{3456}^{(-)}(\mathbf{k},H)$ applies in this region and also using Eq. (7), we find that

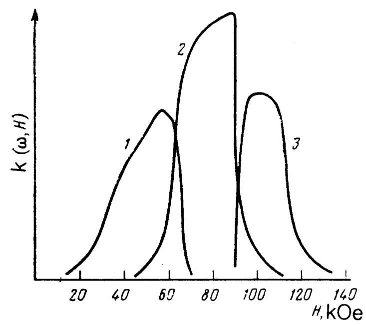

FIG. 7. Theoretical dependence of the two-magnon absorption coefficient of $\omega_{1278}^{(+)}$ exchange magnons on the applied field calculated for different radiation frequencies: 1) $\omega=18\,\mathrm{cm}^{-1}$; 2) $\omega=22\,\mathrm{cm}^{-1}$; 3) $\omega=26\,\mathrm{cm}^{-1}$.

$f^{(\pm)}_{3456}(\mathbf{k})$ is described by

$$f^{(\pm)}_{3456}(\mathbf{k})=\beta\delta^2 H_{AF}\gamma_{13}(\mathbf{k})/4\omega^{(\pm)}_{3456}(\mathbf{k},H). \tag{23}$$

Substituting this quantity into Eq. (22), we shall consider the contributions made to the absorption by the parts of the spin-wave spectrum near various dynamic critical points.

A. Isolated dynamic critical point. In this case, we find that $K^{(\pm)}$ is described by the following expression:

$$\begin{aligned}K^{(\pm)}=&\frac{\pi}{2}K_0|m_\perp^{(\pm)}|\{\arccos\bar{\gamma}_{13}^{(\pm)}\\&-\arccos[\bar{\gamma}_{13}^{(\pm)}+(|m^{(\pm)}|\Delta^2\omega^{(\pm)})^{1/2}]\\&+\bar{\gamma}_{13}^{(\pm)}(1-\gamma_{13}^{(\pm)2})^{1/2}-(\bar{\gamma}_{13}^{(\pm)}+|m^{(\pm)}|\Delta^2\omega^{(\pm)})\\&\times(1-[\bar{\gamma}_{13}^{(\pm)}+|m^{(\pm)}|\Delta^2\omega^{(\pm)}]^2)^{1/2}\}.\end{aligned}\tag{24}$$

Here,

$$\Delta^2\omega^{(\pm)}=\pm\omega^2/4\mp\omega_0^{(\pm)2},\qquad K_0=g^2\beta^2\delta^4 H_{AF}/2\pi\hbar c v_0.$$

The upper sign corresponds to $\omega>2\omega_0^{(+)}$ and the lower to $\omega<2\omega_0^{(-)}$.

B. Nonisolated critical points. In the calculation of $K^{(\pm)}$ we shall now assume that $\cos^2\varkappa_z\approx 1-\varkappa_z^2$. We shall consider just two examples (a more detailed analysis of two-magnon absorption coefficient can be found in Ref. 19).

Two-magnon absorption of the $\omega^{(+)}_{3456}$ mode, when $\omega>2\omega_0^{(+)}$ and $\Delta^2\omega^{(+)}\tilde{m}_\perp^{(+)}\leqslant\varkappa_{0\perp}^4$, is described by

$$K^{(+)}=\pi^2K_0(\tilde{m}_\perp^{(+)}\tilde{m}^{(+)})^{1/2}[1-{}^1/_2\tilde{m}^{(+)}\Delta^2\omega^{(+)}].$$

Two-magnon absorption in the $\omega^{(-)}_{3456}$ mode in the case when $\omega<2\omega_0^{(-)}$ and $|\tilde{m}_\perp^{(-)}|\Delta^2\omega^{(-)}\geqslant\varkappa_{0\perp}^4$ is described by

$$\begin{aligned}K^{(-)}=&2\pi K_0|\tilde{m}_\perp^{(-)}\tilde{m}^{(-)}|^{1/2}\left\{\left(1+\frac{1}{2}\tilde{m}^{(-)}\Delta^2\omega^{(-)}\right)\right.\\&\times\ln\left|\frac{K+(K^2+\tilde{m}^{(-)}\Delta^2\omega^{(-)})^{1/2}}{(\tilde{m}^{(-)}\Delta^2\omega^{(-)})^{1/2}}\right|\\&\left.-\frac{1}{2}K(K^2+\tilde{m}^{(-)}\Delta^2\omega^{(-)})^{1/2}\right\}.\end{aligned}$$

The upper limit to K (the value of which does not affect the frequency dependence of the absorption coefficient which we have to find) can be any value of $\varkappa_z$ which is large compared with the difference $\Delta^2\omega^{(-)}$, but yet sufficiently small so that the expansion of Eq. (18) is still valid.

We shall now note a characteristic feature of the formulas derived above. We can easily see that the contribution made to the two-magnon absorption coefficient by parts of the spectrum located in the direct vicinity of a dynamic critical point contains the parameter $(DJ^{-1})^{1/2}$. This result is to be expected because the very appearance of such critical points is due to the Dzyaloshinskii interaction.

4. TWO-MAGNON ABSORPTION BY EXCHANGE BRANCHES OF THE SPECTRUM OF $CuCl_2\cdot 2H_2O$: EXPERIMENTS

The two-magnon absorption spectra were determined at 2 K using a submillimeter pulsed spectrometer which we employed earlier[12] to study the exchange modes of an antiferromagnetic resonance in the same crystal. Our samples were prepared from $CuCl_2\cdot 2H_2O$ single crystals grown from a saturated solution of copper chloride. The dimensions of these samples were $3\times 3\times 1.5$ mm and their faces were oriented along crystallographic axes.

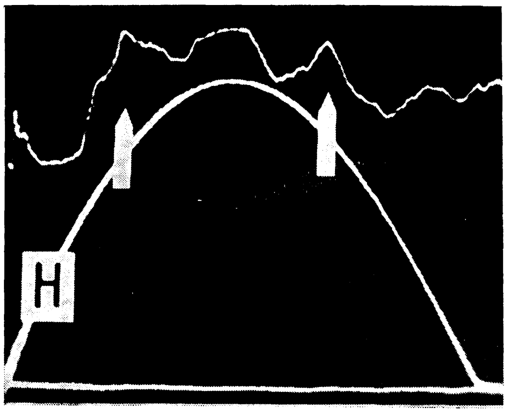

FIG. 8. Spectrogram of the absorption in $CuCl_2\cdot 2H_2O$ at the frequency $\omega=22\ cm^{-1}$. Here, H is the magnetic field signal. The arrows identify the absorption maxima at the leading and trailing edge of the field pulse.

The absorption spectrum was recorded in the range 12–15 cm^{-1} where the weak two-absorption bands were located in the wing of a strong antiferromagnetic resonance line of the acoustic mode $\omega^{(-)}_{3456}$, which made the observations difficult. It should be pointed out that, according to the theoretical calculations, the two-magnon absorption due to dynamic critical points appearing in a crossover situation should appear precisely in this range of frequencies. At higher frequencies 25–30 cm^{-1} there should be two-magnon absorption bands in an external static field, which could not be achieved using our apparatus because of high-voltage breakdown caused by the current terminals of a pulsed solenoid located in a cryostat from which helium was evaporated. It was possible to observe two-magnon absorption bands at frequencies 17–23 cm^{-1}. Calculations of the dispersion laws indicated that we could expect both $\omega^{(+)}_{3456}$ and $\omega^{(+)}_{1278}$ spin waves in this range. However, flattening of the dispersion law of $\omega^{(+)}_{1278}$ spin waves ensured that the intensity of two-magnon absorption in their case was stronger than for $\omega^{(+)}_{3456}$. By way of example, Fig. 8 shows a two-magnon absorption spectrogram for the incident radiation frequency $\omega=22$ cm^{-1} in the presence of an external magnetic field $\mathbf{H}\|\mathbf{a}$ with the $\mathbf{H}_\omega\|\hat{\mathbf{a}}$ polarization. The spectrum was scanned with a pulsed magnetic field. The observed positions of the absorption bands (identified by arrows) were in agreement with the theoretical predictions for the two-magnon absorption by the pure exchange spin wave $\omega^{(+)}_{1278}$. The change in the polarization of the radiation weakened the bands, which was again in agreement with the theory. It should be stressed that, to the best of our knowledge, this was the first observation of two-magnon absorption by exchange magnons. The large difference between the energies of the acoustic $\omega^{(-)}_{1278}$ and exchange $\omega^{(+)}_{1278}$ branches was the reason why the contribution of the states of acoustic magnons to two-magnon absorption by exchange magnons could be ignored.

The observed absorption spectra were asymmetric relative to the leading and trailing edges of the magnetic field pulses. For example, the spectrogram in Fig. 8 had absorption band maxima, identified by arrows, in fields 70 kOe (at the leading edge) and 84 kOe (at the trailing edge). This asymmetry could be due to the heating of a sample by a magnetic field pulse. Moreover, the investigated absorption was of low intensity and the spectra were obtained at the maximum amplification of the spectrometer. Therefore, the experimental results could not be used in a quantitative comparison with the theory and should be regarded as purely preliminary. Our aim is to record the two-magnon absorption spectra using improved apparatus.

1) These linear combinations are the basis functions of the transposition representation of the space symmetry groups of magnetically ordered phases at $\mathbf{k} = 0$.

2) A detailed classification of the components of the irreducible combinations of the spin vectors in accordance with irreducible representations of the group D_{2h}^{7} and an analysis of the ground states can be found in Ref. 12.

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