Exchange modes in antiferromagnetic $CuCl_2 \cdot 2H_2O$

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A systematic experimental and theoretical investigation was made of the exchange modes of a magnetic resonance in a four-sublattice orthorhombic antiferromagnetic $CuCl_2 \cdot 2H_2O$. Measurements were made at a temperature of 1.8 K in the frequency range 6–10 cm^{-1} using magnetic fields up to 10 T applied parallel to the easy magnetization axis. The intensity of the absorption of the exchange modes was directly proportional to the degree of noncollinearity of the magnetic structure, assumed to be collinear in the exchange approximation. A study was made of the region of interaction of the exchange and acoustic modes of identical symmetry and it was found that the parameter describing the coupling between these modes was the anisotropic exchange constant. The ferromagnetic and anisotropic exchange integrals were determined and the magnitude of the bending angle of the sublattices in zero field was found.

I. INTRODUCTION

Most real antiferromagnets have many sublattices. The number of magnetic-resonance modes in such crystals is equal to the number of magnetic sublattices. In the case of crystals in which the exchange interactions are much stronger than the anisotropic interactions the modes in question can be classified in a fairly arbitrary way. The modes with activation energies which vanish in the exchange approximation in the absence of a magnetic field will be called the acoustic modes and the modes with activation energies which remain finite under these conditions will be described as the exchange modes.

This classification reflects the specific features of the precession of the sublattice magnetization participating in the oscillations corresponding to these modes. In the acoustic mode case the precession of the sublattice magnetizations results in small deviation of the "hedgehog of spins" in a unit cell as a whole from a certain special direction. Since this direction is governed by the anisotropic interactions, it follows that the activation energy of the oscillations that correspond to the acoustic modes is governed by the anisotropy. In the case of the exchange mode oscillations the hedgehog of spins is not displaced as a whole, but small deviations of the sublattice magnetizations alter the hedgehog structure. The orientation of the sublattice magnetizations within the hedgehog governs the nature of the magnetic ordering and it is determined by the relationships betwen the sublattice exchange integrals. Therefore, the activation energy of the exchange modes is of the exchange origin.

It is important to stress also that in crystals characterized by a strong single-ion anisotropy of the ions with the spin $S \geqslant 1$ we can expect additional perturbations due to transitions in which the spin projection is not conserved and the energies are of the order of the exchange energy. However, such excitations are not exchange modes because they are not linked to the multisublattice nature of a crystal.

The acoustic modes of an antiferromagnetic resonance (AFMR), the number of which for an antiferromagnetic collinear in the exchange approximation amounts to two, have been investigated both experimentally and theoretically in considerable detail for a large number of crystals with different symmetries. However, such a systematic investigation of the manifestations of the exchange modes in magnetic resonances have not yet been made.

It was reported in Ref. 1 that exchange modes were observed in neutron scattering. Recently, we discovered the exchange modes of $CuCl_2 \cdot 2H_2O$ and carried out a preliminary investigation of the dependences of their frequencies on an external magnetic field.[2] Somewhat earlier the exchange modes were discovered in an $(NH_3)_2(CH_2)_3MnCl_4$ crystal.[3] We also investigated experimentally the field dependences of the frequencies. A confirmation that the absorption lines reported in Ref. 2 were due to the exchange modes was provided by the identity of the dependences of their frequencies and intensities on the magnetic field with those obtained theoretically[4] for a four-sublattice orthorhombic antiferromagnet. Moreover, the behavior of the field dependences of the frequencies and intensities of the exchange modes in the collinear phase reported in Ref. 3 were also in agreement with the predictions of Ref. 4.

The absence of systematic investigations of the exchange modes can be explained by the fact that, firstly, their detection meets with considerable experimental difficulties because the exchange mode frequencies usually lie in the long-wavelength infrared range and the intensities of their absorption lines are low and are governed, as shown below, by the degree of noncollinearity of the magnetic structure. We shall define this degree of noncollinearity as the ratio of the anisotropic exchange interaction (in the Dzyaloshinskiĭ–Moriya sense, as explained in the theoretical part of the present paper) to the isotropic exchange. This ratio governs the angle of misorientation of the sublattice magnetizations within the hedgehog of spins. Secondly, theoretical calcula-

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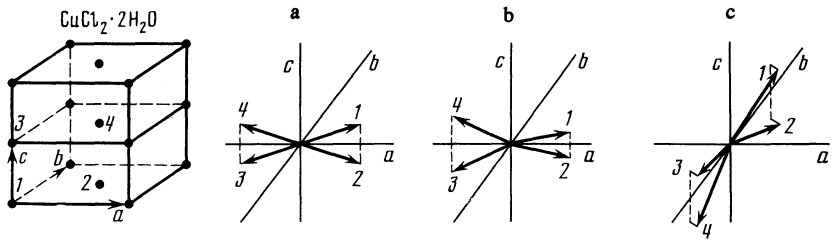

FIG. 1. Unit cell of $CuCl_2 \cdot 2H_2O$ and the ground state in a magnetic field parallel to the **a** axis: a) $H = 0$; b) $H < H_{sf}$; c) $H_{sj} < H < H_f$.

tions of the magnetic resonance spectra of multisublattice magnetic materials are very time-consuming[5] and only the use of calculation methods utilizing symmetry considerations[6] makes it possible to simplify the procedure. For example, the first calculation of the field dependences of the frequencies of the exchange modes in a four-sublattice orthorhombic antiferromagnet was reported by Joenk.[5] However, Joenk studied only the range of fields not exceeding the spin flop transition field on the asumption that the exchange modes could not be observed in the spin-flop phase.

Application of the symmetry approach in Ref. 6 to the study of the field dependences of the frequencies in the AFMR spectrum made it possible to identify those cases in which the exchange and acoustic modes had the same symmetry. As shown below, a strong interaction between the exchange and acoustic modes of the same symmetry is possible in the spin-flop phase if the field is sufficiently strong. An interesting feature of this interaction is that the parameter describing the mode coupling is the anisotropic exchange constant. An investigation of the region of interaction between the exchange and acoustic modes and also (as pointed out above) of the intensities of the absorption lines of the exchange modes should make it possible to determine the anisotropic exchange constant.

A detailed study of AFMR in a multisublattice magnetic material therefore provides an opportunity to obtain quantitative data on the magnetic structure of a crystal, for example, to determine the degree of noncollinearity of the magnetic structure which has hitherto been found only from elastic neutron scattering.

Our aim was to determine in detail the absorption spectrum of the exchange modes in $CuCl_2 \cdot 2H_2O$ crystals, to calculate the corresponding field dependences of the frequencies and characteristics of the absorption lines, and also to find the constants representing the magnetic structure of this crystal within the framework of the four-sublattice model.

THEORY

Copper chloride dihydrate is a compound isomorphous with $CuCl_2 \cdot 2D_2O$, which—according to the neutron diffraction data[7,8]—is a four-sublattice orthorhombic antiferromagnet. The symmetry group of the paramagnetic phase of these crystals is D_{2h}^7. It is shown in Ref. 9, which provides a microscopic justification of the Dzyaloshinskiĭ interaction, that the anisotropic exchange interaction in such antiferromagnets and the associated bending of the magnetization vectors of the sublattices give rise to a weak ferromagnetic moment. However, this weak moment is zero in chloride dihydrate and in $CuCl_2 \cdot 2D_2O$ if there is no external magnetic field. The magnetic structure of $CuCl_2 \cdot 2H_2O$ in the Pbmn setting is shown in Fig. 1. The magnetic cell in these two compounds is equal to twice the crystallochemical cell. The doubling occurs along the **c** axis.

Following the method of calculation of the magnetic resonance frequencies developed in Ref. 6, we shall introduce the following irreducible combinations of the sublattice spins:

$$\begin{aligned} \mathbf{F}&=\mathbf{S}_1+\mathbf{S}_2+\mathbf{S}_3+\mathbf{S}_4, \quad \mathbf{L}_2=\mathbf{S}_1-\mathbf{S}_2+\mathbf{S}_3-\mathbf{S}_4, \\ \mathbf{L}_1&=\mathbf{S}_1+\mathbf{S}_2-\mathbf{S}_3-\mathbf{S}_4, \quad \mathbf{L}_3=\mathbf{S}_1-\mathbf{S}_2-\mathbf{S}_3+\mathbf{S}_4, \end{aligned} \tag{1}$$

which transform in accordance with the irreducible representations of the symmetry group of the paramagnetic phase. A classification of the components of the vectors of Eq. (1) in accordance with irreducible representations of the group D_{2h}^7 is given in Table I. It should be pointed out that the representations $\Gamma_5 - \Gamma_8$ in Table I are the irreducible representations with $\mathbf{q} \neq 0$.

We shall be interested only in the frequencies of a homogeneous resonance for which the Hamiltonian of the system can be written in the form

$$\mathcal{H} = \sum_{ij\alpha\beta} K_{ij}^{\alpha\beta} S_i^{\alpha} S_j^{\beta} - \sum_{ij\nu} g_{ij}^{\nu} S_i^{\nu} H_j,$$

where $i, j = x, y, z$, $\alpha, \beta, \nu = 1, 2, 3, 4$ label the sublattices; $K_{ij}^{\alpha\beta}$ are the constants of the intrasublattice and intersublattice interactions; S_i^{α} is the magnetization component of the α-th sublattice; H is the external magnetic field; g_{ij}^{ν} is the tensor of the g factors of the ν-th sublattice. Using the irreducible representations of Table I, we can rewrite the above Hamiltonian as follows:

$$\begin{aligned} \mathcal{H} = \sum_j \{J_{0j}F_j^2 + J_{1j}L_{1j}^2 + J_{2j}L_{2j}^2 + J_{3j}L_{3j}^2\} + D_1 F_x L_{2z} + D_2 F_z L_{2x} \\ + D_3 L_{1x} L_{3z} + D_4 L_{1z} L_{3x} - H_x(g_{xx}F_x + g_{xz}L_{2z}) \\ - H_y g_{yy} F_y - H_z(g_{zz}F_z + g_{zx}L_{2x}). \end{aligned} \tag{2}$$

TABLE I. Classification of components of combinations of sublattice magnetizations in accordance with irreducible representations of D_{2h}^7 group.

Group representation	Vector components
Γ_1	L_{2y}
Γ_2	F_x L_{2z}
Γ_3	F_y
Γ_4	F_z L_{2x}
Γ_5	L_{3y}
Γ_6	L_{1x} L_{3z}
Γ_7	L_{1y}
Γ_8	L_{1z} L_{3x}

The quantities J and D are defined in terms of K by means of Eq. (1), for example,

$$J_{2y}={}^1/_4(K_{yy}{}^{11}-K_{yy}{}^{12}+K_{yy}{}^{13}-K_{yy}{}^{14}),$$

$$D_3={}^1/_2(K_{xz}{}^{11}-K_{xz}{}^{12}-K_{xz}{}^{13}+K_{xz}{}^{14}).$$

The parameters D_j ($j=1, 2, 3, 4$) relating the off-diagonal (Cartesian indices) components of the irreducible combinations of Eq. (1) will be called the anisotropic exchange interaction constants. These constants describe the Dzyaloshinskiĭ–Moriya interaction. It should be pointed out that the quantities D include a contribution from the symmetric and antisymmetric (in respect of the Cartesian indices) parts of the matrices $K_{ij}^{\alpha\beta}$. As shown in Ref. 9, the symmetric part of the off-diagonal components $K_{ij}^{\alpha\beta}$ ($i\neq j$) is of the order of $(\Delta g/g)^2K_{ii}^{\alpha\beta}$ and the antisymmetric is of the order of $(\Delta g/g)K_{ii}^{\alpha\beta}$, where Δg is the deviation of the g factor from the value of a free electron[9] and $K_{ii}^{\alpha\beta}$ is the intersublattice exchange integral. If these $K_{ii}^{\alpha\beta}$ integrals are of the same order of magnitude or they differ only slightly, the greatest contribution to D is made by the antisymmetric parts of the matrices $K_{ij}^{\alpha\beta}$.

In our case the matrices $K_{ij}^{\alpha\beta}$ for the ion pairs 11 and 13 are symmetric, whereas for the pairs 12 and 14 are antisymmetric, so that we can assume that $K_{xz}^{11}, K_{xz}^{13}\ll K_{xz}^{12}, K_{xz}^{14}$. We shall show later that in the investigation crystal the values of K_{ii}^{13} are only an order of magnitude greater than K_{ii}^{12}. An allowance for the inequalities just given shows that $D_1\approx -D_2$ and $D_3\approx -D_4$.

However, there are some crystals for which the relationships between the intersublattice exchange integrals are such that the constants D are determined by the contributions from the symmetric parts of the matrices $K_{ij}^{\alpha\beta}$ and this is true, for example, of antiferromagnetic $(C_2H_5NH_3)_2CuCl_4$ (Ref. 10).

Since the parameters J include the isotropic exchange interaction, the following relationships should be obeyed[9]

$$|J_{\alpha j}|\gg|D|\gg|J_{\alpha j}-J_{\alpha i}|=A_{\alpha ji}. \tag{3}$$

The quantities $A_{\alpha ji}$ govern the anisotropic interactions.

In the absence of an external magnetic field we can expect a magnetic configuration of copper chloride dihydrate to have finite values of the components L_{1x} and L_{3z} and L_{3z} should be the weak antiferromagnetic component. This magnetic configuration (Fig. 1a) transforms in accordance with the irreducible representation Γ_6 of Table I. We shall now classify the magnetically ordered phases in accordance with the corresponding irreducible representations in Table I, i.e., in the absence of an external magnetic field we shall assume that the magnetic structure of $CuCl_2\cdot 2H_2O$ is the ordered Γ_6 phase.

The distribution of the magnetizations in a hedgehog of spins shown in Fig. 1a corresponds to the L_1-type antiferromagnetic ordering, i.e., in the exchange approximation the only nonzero quantity is the principal antiferromagnetic vector $\mathbf{L}_1$. The absence of the L_1 ordering means that the quantities occurring in Eq. (2) satisfy the relationships

$$|J_{1x}|=-J_{1x}>-J_{1y},\ -J_{1z}>|J_{0i}|,\ |J_{2i}|,\ |J_{3i}|. \tag{4}$$

It is quite clear that, depending on the values of the parameters J and D, and also on the magnitude and direction of the magnetic field in a given structure, we can encounter also other types of magnetic ordering listed in Table I. The set of ground states which may be assumed by copper chloride dihydrate in an external magnetic field oriented along the **a** axis is shown in Fig. 1 [the coordinate system x, y, z selected in Eq. (2) coincides with the **a**, **b**, and **c** axes].

A magnetic field $\mathbf{H}\|\mathbf{a}$ induces a finite value of F_x, which gives rise to L_{2z}. Consequently, a mixed Γ_{26} magnetic configuration is established and it correspond to the following equilibrium values of the irreducible combinations of Eq. (1):

$$L_{1x}\approx 4S,\quad L_{2z}\approx {}^1/_4HD_3(J_{3z}-J_{1x})^{-1}(J_{2z}-J_{1x})^{-1},$$
$$L_{3z}\approx -2SD_3(J_{3z}-J_{1x})^{-1},\quad F_x\approx {}^1/_8D_3H(J_{3z}-J_{1x})^{-2}(J_{2z}-J_{1x})^{-1}. \tag{5}$$

The equilibrium values of the vectors in Eq. (1) can be found by the method described in Ref. 6 or using the Lagrange multipliers.

Here and later we shall ignore the quantities g_{xz} and g_{zx}, which are usually small. Throughout the range of fields corresponding to the Γ_{26} phase we have $F_x\ll L_{1x}$. Bearing in mind the relationship $|J_3-J_1|\propto D^2$ (Ref. 9), we can estimate F_x from $F_x\propto HJ^{-2}$. It should be pointed out that the constants D and J in Eq. (2) are dimensionless and the value of S has the dimensions of magnetization.

The expressions for the equilibrium values of the irreducible combinations for the phase Γ_6 are obtained from Eq. (5) when $H=0$. It readily follows from the expressions in Eq. (5) and from Fig. 1a that if $H=0$, then the degree of noncollinearity of the magnetic structure is governed by the rate of the weak antiferromagnetic component to the principal antiferromagnetic vector $|L_{3z}L_{1x}^{-1}|=2D_3(J_{3z}-J_{1x})^{-1}$. The lability field of the collinear phase is given by the expression

$$H_\parallel{}^2=H_{E0y}H_{A1y},\quad H_{E\mu i}=8S(J_{\mu i}-J_{1x}),\quad \mu=0, 2, 3;$$
$$H_{A1y}=8S[J_{1y}-J_{1x}+{}^1/_4D_3{}^2(J_{3z}-J_{1x})^{-1}]. \tag{6}$$

When the fields exceed the spin-flop field of the magnetic moments of the sublattices, the principal antiferromagnetic vector is parallel to the **b** axis and the magnetically ordered phase is Γ_{27}, for which we have

$$L_{1y}=4S(1-H^2H_f^{-2})^{1/2},\quad L_{2z}=-16S^2D_1HH_{\Delta 2z}^{-1}H_f^{-1}, \tag{7}$$
$$F_x=4SHH_f^{-1},\ H_f=H_{\Delta 0x}-(4SD_1)^2H_{\Delta 2z}^{-1},\quad H_{\Delta\mu i}=8S(J_{\mu i}-J_{1y}).$$

In the spin-flop phase the weak antiferromagnetic vector L_{2z} increases on increase in the magnetic field, but even in fields close to the exchange value $H\approx H_f$ it remains a small quantity (of the order of DJ^{-1}) compared with $F_x=4S$. This spin-flop phase is stable in fields $H_\perp<H<H_f$, where

$$H_\perp{}^2=H_f{}^2H_{A1y}[H_{\Delta 0y}+H_{A1y}]^{-1}. \tag{8}$$

The spin flopping of the magnetic moments of the sublattices, i.e., the transition from the Γ_{26} to the Γ_{27} phase, is a first-order phase transition in $CuCl_2\cdot 2H_2O$.

In a magnetic field $H=H_f$ the phase transition caused

TABLE II. Classification of antiferromagnetic resonance frequencies of $CuCl_2 \cdot 2H_2O$ in accordance with symmetry (EM and AM are exchange and acoustic modes).

Symmetry of magnetic ordering	Symmetry of homogeneous oscillations	Number of resonance modes
Γ_6 ($H=0$)	Γ_{16}	1 EM
	Γ_{25}	1 EM
	Γ_{38}	1 AM
	Γ_{47}	1 AM
Γ_{26} ($H<H_{sf}$)	Γ_{1256}	2 EM
	Γ_{3478}	2 AM
Γ_{27} ($H_{sf}<H<H_f$)	Γ_{1278}	1 EM + 1AM
	Γ_{3456}	1 EM + 1AM

collapse of the magnetic moments of the sublattices, which causes the principal antiferromagnetic vector L_{1y} to vanish, in accordance with Eq. (7). However, the weak antiferromagnetic vector L_{2z} remains, as already pointed out, different from zero.

An interesting feature of this sublattice collapse transition in $CuCl_2 \cdot 2H_2O$ is the simultaneous disappearance of the doubling of the magnetic unit cell. It readily follows from Fig. 1 and from the expressions in Eq. (7) that in the field $H = H_f$ the directions of the spins of the ions 1, 3, and 2, 4 coincide and the magnetic structure of the crystal in the collapsed phase, i.e., in fields $H > H_f$, should be described by the two-sublattice model.

We shall now consider expressions for the exchange and acoustic modes of a magnetic resonance in a four-sublattice orthorhombic antiferromagnet. Following Ref. 6, the AFMR modes can be conveniently classified in accordance with the irreducible representations of the paramagnetic phase group. The essence of this classification is that we can identify the small deviations from the irreducible combinations of Eq. (1) that participate in oscillations representing a given magnetic mode. It is shown in Ref. 11 that a direct product of an irreducible representation of the paramagnetic phase group, which governs the transformations of small deviations of the irreducible combinations of Eq. (1) corresponding to one magnon mode, always includes the irreducible representation of the appropriate magnetically ordered phase. Table II gives a classification of all the AFMR modes of $CuCl_2 \cdot 2H_2O$ in a magnetic field applied parallel to the **a** axis.

The separation of the modes into exchange and acoustic is made in accordance with the following criterion. Since the acoustic modes correspond to a rotation of the hedgehog of spins as a whole, it follows that they are the modes to which the irreducible representations of small deviations of the principal antiferromagnetic vector apply. For example, in the case of the Γ_6 phase these are the small deviations of the vector L_1, i.e., L_{1z} (irreducible representation Γ_8) and L_{1y} (irreducible representation Γ_7). However, in the case of the Γ_{27} phase, the corresponding vectors are L_{1x} and L_{1z} with the irreducible representations Γ_6 and Γ_8, respectively.

The condition for the excitation of a given AFMR mode is the presence in its oscillations of small deviations of the vector **F**. For example, in the Γ_6 phase the ν_{16} mode is not excited because no component of the vector **F** transforms in accordance with the representations Γ_1 and Γ_6 and the ν_{25} mode may be excited only when the polarization of the high-frequency magnetic field is parallel to the x axis.

It is clear from Table II that in the Γ_{27} phase the exchange and acoustic modes have the same symmetry, i.e., oscillations corresponding to an exchange mode include a contribution from small deviations of the principal antiferromagnetic vector. However, as shown in Ref. 11, in the exchange branches of the AFMR spectrum the largest amplitude of the oscillations is exhibited by the weak antiferromagnetic vector, whereas in the acoustic modes this is true of the principal antiferromagnetic vector.

In the collinear phase (Γ_{26}) the AFMR frequencies are given by the expressions

$$\nu_{1256}(\pm) = \frac{g}{\sqrt{2}}\{H_{e1}^2+H_{e2}^2+2H^2 \pm R_e\}^{1/2}, \tag{9}$$

$$\nu_{3478}(\pm) = \frac{g}{\sqrt{2}}\{H_{a1}^2+H_{a2}^2+2H^2 \pm R_a\}^{1/2},$$

where

$$R_e = \{(H_{e1}^2+H_{e2}^2+2H^2)^2 - 4(H_{e3}^2-H^2)(H_{e4}^2-H^2)\}^{1/2},$$

$$R_a = \{(H_{a1}^2+H_{a2}^2+2H^2)^2 - 4(H_{a3}^2-H^2)(H_{\parallel}^2-H^2)\}^{1/2},$$

$$H_{e1}^2 = H_{E3z}H_{E2y},\ H_{e2}^2 = H_{E3y}H_{E2z},\ H_{e3}^2 = H_{E3z}H_{E2z},\ H_{e4}^2 = H_{E3y}H_{E2y},$$

$$H_{a1}^2 = H_{E0y}H_{A1z},\ H_{a2}^2 = H_{E0z}H_{A1y},\ H_{a3}^2 = H_{E0z}H_{A1z},$$

$$H_{A1z} = 8S(J_{1z}-J_{1x}).$$

The lower index for the frequencies in the system (9) correspond to the irreducible representation in Table II which governs the transformation of oscillations of this frequency. In the Γ_{26} collinear phase the oscillations corresponding to the exchange and acoustic modes have different symmetries.[4] Therefore, in this phase the existence of the exchange modes has no influence on the positions and behavior of the acoustic modes. However, in low fields $H \ll H_{e1} - H_{e2} (H \ll H_{sf})$ the difference between the exchange mode frequencies varies with the magnetic field proportional to H^2. On the other hand, in the interval of fields $H_{e1} - H_{e2} \ll H \leqslant H_{sf}$ the frequencies of the exchange modes depend linearly on the field.

In the spin-flop phase Γ_{27} the oscillations corresponding to the exchange and acoustic modes have then pairwise-equal symmetries.[4] The dispersion equation governing the AFMR frequencies for this phase is governed by a twelfth-order determinant which splits into two blocks corresponding to pairs of modes of the same symmetry. If we now assume that the values of D_i vanish, each of these blocks splits again into two. Therefore, the constants describing the anisotropic exchange are the "parameters of the coupling" of the exchange and acoustic oscillation modes of the same frequency. The existence of this coupling results in a strong interaction of the oscillations in the part of the spectrum where the frequencies of the exchange and acoustic modes are similar. This interaction suppresses crossing of the field dependences of the exchange and acoustic mode frequencies even when the magnetic field was oriented exactly along the **a** axis. The minimum separation between the frequencies of the characteristic field $H = H_m$ (H_m is deduced from the condition of coincidence of the exchange and acoustic mode

frequencies when $D=0$) is proportional to the constant D.

In fields $H \gtrsim H_{sf}$ and $H \lesssim H_f$ the dependences of the frequencies on the field are described by

$$\begin{aligned}
&\nu_{3456}^2(+)=\nu_{0+}^2 \approx (g/4S)^2[H_{\Delta 3z}H_{\Delta 2y}L_{1y}^2+H_{\Delta 3z}H_{\Delta 3y}F_x^2],\\
&\nu_{3456}^2(-)=\nu_{0-}^2 \approx (g/4S)^2[H_{\Delta 0y}H_{\Delta 0z}F_x^2-H_{\Delta 0z}H_{A1y}L_{1y}^2],\\
&\nu_{1278}^2(+) \approx (g/4S)^2[H_{\Delta 2z}H_{\Delta 3x}L_{1y}^2+H_{\Delta 2z}^2F_x^2],\\
&\nu_{1278}^2(-) \approx (g/4S)^2 H_f H_{A1z}L_{1y}^2,
\end{aligned} \tag{10}$$

where $H'_{A\,1z}=8S\left[J_{1z}-J_{1y}-\frac{1}{4}D_3^2(J_{3x}-J_{1y})^{-1}\right]$.

Solutions of the system (10) were obtained for the case when $\nu_+ \gg \nu_-$. For the $\nu_{1278}(\pm)$ modes this condition is satisfied throughout the investigated range of magnetic fields right up to the sublattice collapse field, whereas in the case of the $\nu_{3456}(\pm)$ modes there is a range of fields where $\nu_{3456}(+) \approx \nu_{3456}(-)$.

We shall consider this region of a strong interaction between the exchange and acoustic modes in greater detail. The results can be presented in a clear manner if we make a number of simplifying assumptions. We have mentioned above that the greatest contribution to the value of D comes from the antisymmetric parts of the intersublattice exchange integrals of the pairs of ions 12 and 14. The contribution made to the exchange integral by the pairs of ions 14 can be ignored because they are more distant than the pairs 12. We therefore obtain $D_1=-D_2=D_3=-D_4=D$. We shall also ignore the exchange integral of the pairs 14 in the expressions for the characteristic fields $H_{\Delta 0}$, $H_{\Delta 2}$, and $H_{\Delta 3}$. Moreover, we shall neglect the anisotropy of these exchange fields so that $H_{\Delta i}=H_{Ei}$ and $H_{\Delta 2}=H_{\Delta 0}+H_{\Delta 3}$. A solution of the dispersion equation for the frequencies $\nu_{3456}(\pm)$ gives (we shall drop the identified index of the frequency)

$$\nu^2(\pm)=\frac{1}{2}\left\{\nu_{0+}^2+\nu_{0-}^2 \pm \left[(\nu_{0+}^2-\nu_{0-}^2)^2 + \frac{g^4}{(2S)^2}D^2L_{1y}^2[H_{\Delta 2}^2L_{1y}^2-(H_{\Delta 0}^2-H_{\Delta 2}H_{\Delta 3})F_x^2]\right]^{1/2}\right\}. \tag{11}$$

In Eq. (10), the frequency ν_{0-} is described by an expression for the characteristic field $H_{A\,1y}$, with $D=0$. Directly in a field H_m for which we have $\nu_{0+}=\nu_{0-}=\nu(H_m)$, we obtain

$$\nu(\pm)=\nu(H_m)\pm\frac{g^2}{8S}DL_{1y} \times [H_{\Delta 2}^2L_{1y}^2-(H_{\Delta 0}^2-H_{\Delta 2}H_{\Delta 3})F_x^2]^{1/2}\nu^{-1}(H_m). \tag{12}$$

The magnitude of the smallest separation between the frequencies of the interacting modes is

$$\Delta\nu_{min}=\frac{g^2}{4S}DL_{1y}[H_{\Delta 2}^2L_{1y}^2-(H_{\Delta 0}^2-H_{\Delta 2}H_{\Delta 3})F_x^2]^{1/2}\nu^{-1}(H_m). \tag{13}$$

It follows from the above expression that the constant D can be found by measuring the smallest separation between the frequencies. However, in the range of weak fields $H \lesssim H_{sf}$ in the case of the Γ_{26} phase and fields $H \gtrsim H_{sf}$ in the case of Γ_{27} phase the anisotropic exchange does not affect in any way the behavior of the acoustic modes of AFMR. It is clear from Eqs. (9) and (10) that the constant D simply renormalizes the anisotropy field.

EXPERIMENTAL METHOD

Measurements were made by the rf method in the frequency range 6–10 cm^{-1} using a sweep-type spectrometer.[12] The sources of microwave radiation were backward-wave tubes and a crystal of n-type InSb cooled to 4.2 K served as a detector. The wavelength was measured by an interference wavemeter to within 0.2%. The wavelength stability was governed mainly by the stability of the voltage using a delay system of a backward-wave tube (0.01%), which was monitored using a digital voltmeter.

The conditions for the excitation of the exchange modes in the collinear phase and of the $\nu_{1278}(+)$ mode in the spin-flop phase (with the magnetic vector of the microwave radiation oriented so that $\mathbf{h}\|\mathbf{H}\|\mathbf{a}$) were satisfied employing a pulsed Helmholtz solenoid (Fig. 2a). Microwave radiation traveled along the **b** axis of the sample and the orientation of **h** could be varied in the **ac** plane. Measurements in the region of the interaction between the exchange and acoustic modes $\nu_{3456}(\pm)$ were made with the microwave radiation polarized so that $\mathbf{h}\|\mathbf{c}$ and in this case a conventional pulsed solenoid was used (Fig. 2c). The radiation traveled along the **a** axis and the polarization **h** was established in the **bc** plane.

A sample was placed in a solenoid channel between two Teflon quasioptic waveguides matched to a microwave channel by cone–lens junctions. The measuring cell of the spectrometer, described in detail in Ref. 12, ensured that the polarization was not affected and that the losses were low in a wide range of wavelengths. The intensity of the magnetic field created by the pulsed solenoids was deduced from the integrated signal produced by a detection coil wound on the measuring cell waveguide at the point of location of the sample. The field signal was calibrated using the AFMR line of $RbMnF_3$. A polyethylene pellet containing a small amount of this substance was placed alongside the sample. The zero level of the field signal was recorded automatically for every magnetic field pulse, so that the long-term instability of the parameters of the recording circuit did not affect the experimental error. Consequently, the error in the determination of the absolute value of the magnetic field did not exceed 1 – 2%.

In these experiments we used optically homogeneous $CuCl_2 \cdot 2H_2O$ single crystals grown from a saturated solution of pure copper chloride. The samples were in the form of a cube with an edge of 3 mm and with faceting along the crystallographic axes. In an investigation of the region of the interaction between the exchange and acoustic modes we ensured that an undistorted AFMR line was obtained (its intensity far from the interaction region was 500 times higher than the intensity of the exchange mode line) by employing a sample 0.35 mm thick and of 3×3 mm^2 area with the **a** axis perpendicular to the plane of the plate. Orientation of a sample along two mutually perpendicular planes was set on the basis of the maximum value of the splitting of the exchange mode frequencies in fields $H \approx H_{sf}$ [it was found that $CuCl_2 \cdot 2H_2O$, like MnF_2 (Ref. 13), exhibited field dependences of the magnetic resonance frequency which at

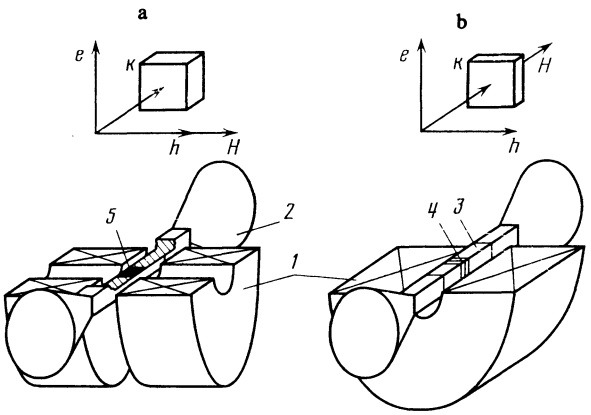

FIG. 2. Experimental geometry and schematic representation of the cells used in the measurements: a) pulsed Helmholtz solenoid; b) pulsed conventional solenoid. The numbers have the following meaning: 1) solenoid winding; 2) quasioptic waveguide with a core–lens junction; 3) waveguide support; 4) detection coil for measuring the magnetic field; 5) investigated sample inside support 3; **k**, **e**, and **h** are the directions of the wave, electric, and magnetic vectors of microwave radiation.

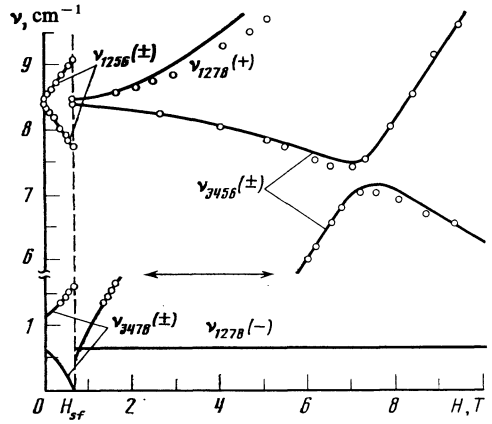

FIG. 3. Field dependences of the AFMR frequencies of $CuCl_2 \cdot 2H_2O$.

$H \approx H_{sf}$ depended strongly on the orientation of a sample in a magnetic field]. The orientation **H**||**a** was set in the **ab** plane, where the sensitivity to the orientation was strongest, to within $\pm 3'$.

In spite of the fact that we used fairly thick samples (3 mm) the absorption of the exchange mode was only 5 – 10%. Convenient spectrograms were obtained by increasing the gain of the absorption signal channel in a calibrated manner, so that the precision of the absorption intensity measurements was retained.

A working temperature of about 1.8 K was achieved by pumping out helium vapor from a cryostat where a pulsed solenoid with the investigated sample was located. The Néel temperature of $CuCl_2 \cdot 2H_2O$ is known to be 4.33 K.

DISCUSSION OF EXPERIMENTAL RESULTS

Preliminary measurements of the field dependences of the exchange mode frequency in $CuCl_2 \cdot 2H_2O$ crystals[8] were carried out with the axis tilted at an angle of about 5° relative to the external magnetic field. This tilt was greater than the critical angle for the first-order phase transition, which amounted to 20′ for $CuCl_2 \cdot 2H_2O$ (Ref. 14). Consequently, it was reported in Ref. 2 that in fields $H \approx H_{sf}$ the dependence of the exchange mode frequency on the magnetic field was continuous. In the present study these measurements were repeated but with H inclined to the **a** axis by an angle not exceeding $\pm 3'$. When this precision of the orientation was achieved, the experimental field dependence of the exchange mode frequency (Fig. 3) exhibited discontinuities at $H \approx H_{sf}$. By way of example, we plotted in Fig. 4a an absorption spectrogram at 8.6 cm^{-1}. In a field $H \approx H_{sf}$ there was practically no absorption. The absorption lines observed in the same spectrogram corresponded to the $\nu_{1256}(+)$ oscillations (in lower fields) and to the $\nu_{1278}(+)$ oscillations (in higher fields). In zero field the frequencies $\nu_{1256}(\pm)$ were $\nu_{16} = 8.45$ cm^{-1}, respectively.

The continuous curves in Fig. 3 are the results of a theoretical calculation. This calculation was carried out employing the approximation discussed above in the theoretical part of the paper. An allowance for a finite temperature was reduced to an allowance for the temperature dependence of the sublattice magnetizations by the molecular field method. When the anisotropy fields were ignored, the exchange mode frequency in zero field was $\nu_0 = g(H_2H_3)^{1/2}$. The fields H_3 and H_2 were governed by the ferromagnetic exchange integrals $J_F \propto K$ [12] and by the sum of the ferromagnetic and antiferromagnetic exchange integrals $J_A \propto K$ [13], respectively. The antiferromagnetic exchange integral determined the field in which the sublattices collapsed and which, at $T = 1.1$ K, was $H_{\Delta 0} = 15$ T (Ref. 15). If we assumed that $H_2 = H_{\Delta 0} + H_3$ and that the half-sum of the exchange mode frequencies in $H = 0$ was $\nu_0 = 8.425$ cm^{-1}, we found that $H_3 = 3.65$ T. Moreover, we assumed that $g = g_0 \mu_B h^{-1}$, where h is the Planck constant, μ_B is the Bohr magneton, and $g_0 = g_{xx} = 2.187$. The ratio of the ferromagnetic and antiferromagnetic exchange integrals found allowing for the interaction only between the nearest neighbors is

$$J_F J_A^{-1} = {}^1/_2 H_3 H_{\Delta 0}^{-1} = 0{,}121,$$

in good agreement with the results of Refs. 15 and 16 obtained from experiments carried out in very high fields.

The exchange modes of the collinear phase and also the exchange modes $[\nu_{1278}(+)]$ in fields exceeding H_{sf} were excited when the microwave radiation polarization was **h**||**a**, whereas the $\nu_{3456}(\pm)$ modes in the spin-flop phase were excited in the **h**||**c** polarization. These conditions for the observation of the exchange modes were in agreement with the results of a theoretical calculation of the dynamic susceptibility of a four-sublattice orthorhombic antiferromagnet,[4] according to which the intensity of the exchange mode $\nu_{3456}(+)$ in the spin-flop phase should be less for the **h**||**b** polarization than for **h**||**c**. By way of illustration, we shall give the expressions for $\chi_{yy}(\nu)$ and $\chi_{zz}(\nu)$ in the spin-flop phase:

$$\chi_{yy}(\nu) = \frac{g^2 F_x H}{\nu_{3456}^2(-) - \nu^2} - \frac{D^2 L_{1y}^2}{\nu_{3456}^2(+)} \frac{g^4 F_x H}{\nu_{3456}^2(+) - \nu^2}, \qquad (14)$$

$$\chi_{zz}(\nu) = \frac{F_x H^{-1} \nu_{3456}^2(-)}{\nu_{3456}^2(-) - \nu^2} + \frac{D^2}{2(J_{3x} - J_{1y})} \frac{g^2 L_{1y}^2}{\nu_{3456}^2(+) - \nu^2}.$$

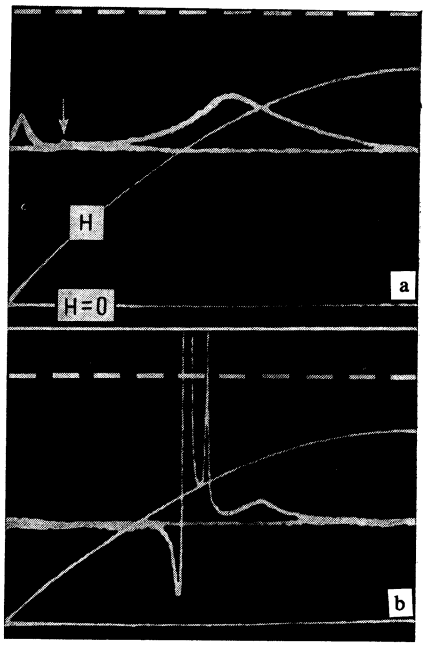

FIG. 4. Absorption spectrograms: a) frequency 8.6 cm^{-1}, H is the magnetic field signal, $H = 0$ is the zero level, the arrow identifies the region of H_{sf}, and the dashed line is the absorption level of 20%; b) frequency 6.75 cm^{-1}. The middle narrow absorption line is the AFMR line of $RbMnF_3$ used for calibration.

It is quite clear from the above formulas that if $\mathbf{h}\|\mathbf{b}$ then the residue of $\chi_{ij}(\nu)$ for the exchange mode contains the factor D^2J^{-2}, whereas for $\mathbf{h}\|\mathbf{c}$ the residue contains the factor D^2J^{-1}. Our experiments, carried out in the $\mathbf{h}\|\mathbf{b}$ configuration, indicated that the mode in question was not observed.

The explicit form of the components of the tensor $\chi_{ij}(\nu)$ demonstrated that the low intensity of the exchange mode lines was due to the fact that the residues $\chi_{ij}(\nu)$ corresponding to these modes always included the degree of noncollinearity of the magnetic structure as a factor. This result is common to all the many-sublattice magnetic materials for which the exchange approximation is valid, irrespective of their symmetry. The absorption lines corresponding to the exchange modes have a profile intermediate between Lorentzian and Gaussian. Measurements of the intensities of the exchange mode lines in fields $H < H_{sf}$ demonstrated that for $H = 0$, the intensity at the maximum of the $\nu_{1256}(-)$ exchange mode was 0.16 ± 0.02 cm^{-1}, whereas in the case of the $\nu_{1256}(+)$ mode it was 0.04 ± 0.02 cm^{-1}. The nonzero intensity of the $\nu_{1256}(+)$ exchange mode in $H = 0$ [calculations of $\chi_{ik}(\nu)$ reported in Ref. 4 indicated that in $H = 0$ this mode should not be excited by an alternating field] could be explained by a nonlinear interaction of the exchange mode oscillations that appear because of the proximity of their frequencies in zero field. On increase in the external magnetic field to (0.1–0.2) T the intensities of the exchange modes become equal within the limits of the experimental error (in fact, this occurred within the limits of the absorption line width). In a field $H = 0.3$ T the intensities at the maxima were 0.1 ± 0.02 cm^{-1}, the width at the half-maximum was 0.07 ± 0.015 cm^{-1}, and the integrated intensity was $(5 \pm 2) \times 10^{-3}$ cm^{-2}. The dependence of the exchange mode frequencies on the external field in the range $H < H_{sf}$ was linear. The nonlinearity of this dependence predicted by the calculations of Ref. 4 and the changes in the intensities of the exchange mode lines in weak fields were clearly significant only within the limits of the absorption line width and could not be detected experimentally.

In fields $H = 7$ T we observed an interaction between the $\nu_{3456}(\pm)$ exchange and acoustic modes when they approached each other. Since these modes had the same symmetry, the interaction of the field dependences of the frequency was not observed even when the axis **a** of the sample was oriented exactly along the external magnetic field (Fig. 3). When the lines approached each other, there was a characteristic transfer of the intensity from the stronger acoustic AFMR mode to the weaker exchange mode. In a field of $H = 6.9$ T there were two absorption lines with identical intensities equal to half the intensity of the acoustic mode line far from the interaction region. By way of example, demonstrating the increase in the intensity of the exchange mode and the reduction in the influence of the degree of noncollinearity, we shall give the values of the residues f_{zz} of the components of the high-frequency susceptibility χ_{zz}:

$$f_{zz}^{(\pm)} = \mp \frac{g^2}{4S} \frac{(\nu^2(\pm) - \nu_{0+}^2) H_{\Delta 0} F_x^2 + (gDL_{1y}/4S)^2 [(H_0 - H_3) F_x^2 - H_2 L_{1y}^2]}{\{(\nu_{0+}^2 - \nu_{0-}^2)^2 - (g^2 DL_{1y}/2S)^2 [H_2^2 L_{1y}^2 - (H_0^2 - H_3 H_2) F_x^2]\}^{1/2}},$$

from which it follows that if $\nu_{0+} = \nu_{0-}$, then

$$f_{zz}^{(+)} = -f_{zz}^{(-)} = -\frac{g^2}{8S} H_0 F_x^2.$$

A further increase in the field causes the intensity of the exchange mode line of frequency given by the value ν_- in Eq. (11) to fall on reduction in L_{1y}^2 [see residues in Eq. (14)]. In fields $H > 10$ T this mode is practically unobservable. The behavior of the lines in the interaction region can be judged on the basis of Fig. 4b, which shows an absorption spectrogram 6.75 cm^{-1}. A strong line with a cutoff top (because of an increase in the gain of the signal channel) corresponds to the angular mode $\nu_{3456}(-)$. The weak line on the right represents absorption at the exchange mode frequency. The narrow peak in the right-hand wing of the acoustic mode line represents AFMR in $RbMnF_3$.

The anisotropic exchange interaction constant D, which is the "coupling parameter" of the exchange and acoustic modes, was found from the condition of matching of the experimental and theoretical values of the lowest frequency difference (13) between the interacting modes in the vicinity of the fields H_m. As established experimentally, this difference varies weakly when temperature is increased to 3 K and the field is inclined to the **a** axis within the limits of $\pm 1°$; the difference amounts to $\Delta\nu_{min} = 0.4 \pm 0.1$ cm^{-1}. The value $4SD = 0.25 \pm 0.06$ T makes it possible to calculate the ratio of the weak antiferromagnetic component to

the principal antiferromagnetic vector and, consequently, to find the tangent of the angle of bending of the sublattices inside the hedgehog of spins in zero field is

$$|L_{3z}L_{1x}^{-1}|=4SDH_3^{-1}=\text{tg}\,\alpha=0.07.$$

This value is in good agreement with the results of neutron diffraction studies of $CuCl_2 \cdot 2D_2O$ (Ref. 8), which is isomorphous with $CuCl_2 \cdot 2H_2O$ and for which the value 0.062 has been obtained.

CONCLUSIONS

The compound $CuCl_2 \cdot 2H_2O$ was used as an example of a four-sublattice orthorhombic antiferromagnetic and an experimental as well as a theoretical study was made of the exchange AFMR modes and the interaction between the exchange and acoustic modes of the same symmetry. These experimental results confirmed well the predictions of the theory and can be used to obtain quantitative information on the magnetic structure of a crystal.

In spite of the fact that the calculations and experiments described above were carried out for the four-sublattice antiferromagnet $CuCl_2 \cdot 2H_2O$, all the characteristic features of the manifestation of the exchange modes in the AFMR spectrum are retained also in the case of the orthorhombic antiferromagnets of different types. For example, it follows from an analysis of the components of the tensor $\chi_{ij}(\nu)$ that the low intensity of the absorption lines of the exchange modes is due to the weak noncollinearity of the magnetic structure of a crystal, because the residues $\chi_{ij}(\nu)$ corresponding to these modes always contain it as a factor. In particular, if the antiferromagnetic exchange integral is larger than the ferromagnetic integral,[1)] then in the spin-flop phase there is always a range of magnetic fields in which the exchange and acoustic mode frequencies of the same symmetry become of the same order of magnitude and a strong interaction occurs between them.

In those cases when the ferromagnetic exchange integral is much less than the antiferromagnetic exchange integral, the frequencies of the exchange modes may be of the order of the acoustic mode frequencies even in fairly low magnetic fields. Such a situation is reported in Ref. 3. Another example of a crystal with the anomalous ratio of the antiferromagnetic and ferromagnetic exchange integrals is the four-sublattice compound $(C_2H_5NH_3)_2 \cdot CuCl_4$, characterized by $J_F \gg J_A$ (Ref. 10). Although in the spin-flop phase of this antiferromagnet all four AFMR modes have the same symmetry, the frequencies of the exchange modes are considerably higher than the frequencies of the acoustic modes throughout the investigated range of fields right up to the value at which the sublattice collapse takes place and the interaction between the exchange and acoustic modes is no longer manifested.[17]

We shall now consider the conditions of validity of the two-sublattice model used to describe the high-frequency properties of multisublattice antiferromagnets, because this model is now generally accepted. It is clear from the above discussion that there is also a closely related problem of the interaction between the exchange and acoustic modes. For example, if the exchange interaction in any antiferromagnetic is unsuitable, i.e., if the relativistic interactions are of the same order of magnitude as the exchange interactions, then the division into the exchange and acoustic modes is no longer meaningful. The energies of all these modes in a multisublattice antiferromagnet should be of the same order of magnitude and, therefore, oscillations of the same symmetry may be subject to a strong interaction. In this situation it is not possible to use the two-sublattice model.

In the case of a multisublattice antiferromagnet in which for some reason or another the anisotropic exchange is weak or absent, we can use a two sublattice model to describe correctly the behavior of the acoustic modes throughout the investigated range of magnetic fields.

However, the situation considered by us above corresponds to intermediate values of the anisotropic exchange constant. There are intervals of magnetic fields in which the two-sublattice model describes well the behavior of the acoustic modes, for example, near the spin-flop transition field. This is due to the fact that if the order parameters of a spin-reorientation transition in a multisublattice magnetic material are the components of the antiferromagnetic vector (i.e., are a linear combination of the sublattice spins in strongest and zero fields, which determine the type of ordering), whereas the Landau expansion in terms of the order parameter near the transition point in fact corresponds to a description of an antiferromagnet in the two-sublattice model. Therefore, the behavior of the magnon mode which becomes softer somewhere in the region of the transition in a multisublattice magnetic material should also be described correctly by this model.

In this sense the separation (on the basis of symmetry considerations) of the order parameter and a gradual establishment of the free energy of the material allowing for its symmetry[18] has made it possible to reduce strongly the number of sublattices and to give a correct qualitative and sometimes quantitative pattern of the behavior of the static properties of a multisublattice magnetic material when, for example, temperature, magnetic field, and other thermodynamic parameters are varied.

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[1)]We are speaking here of the ferromagnetic exchange between inequivalent sublattices. In our case these are the sublattices 1 and 2.

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