# Two-magnon absorption of electromagnetic waves in the exchange noncollinear antiferromagnet $Nd_2CuO_4$

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The two-magnon absorption of electromagnetic waves in the noncollinear magnetic phase of the four-sublattice antiferromagnet $Nd_2CuO_4$ has been considered. The crystal is shown to exhibit a distinguishing feature that the electric dipole moment associated with the pair of spins has an exchange nature. Consequently, the two-magnon absorption of the electromagnetic waves is electrodipole active in the exchange approximation. We have studied theoretically both the magnetic-dipole and electric-dipole channels of the two-magnon absorption and analyzed the frequency dependences of the coefficient of the absorption. The intensities of the absorption processes involving the exchange magnons are shown to be of the same order of magnitude as that involving the acoustic magnons only.

## I. INTRODUCTION

A theoretical analysis of the two-magnon absorption of electromagnetic waves in ferromagnets has been carried out in Ref. 1. The microscopic mechanism of this phenomenon consists in the decay of the quantum of electromagnetic radiation into two magnons. It follows from the laws of conservation of energy and momenta for this process that the wave vector of light may be considered as negligibly small. It means that the absorption process gives rise to magnons with opposite wave vectors corresponding to different points in the Brillouin zone. At the same time the combined energy of the magnons is fixed and equal to the energy of the photon. The absorption intensity is directly proportional to the density of the magnon states. That is, the absorption will be higher for those regions of the spectrum which correspond to the flattened portions of the curves describing the dependences of the magnon energies on the wave vector. It is apparent that quasi-one- and quasi-two-dimensional magnets are the most suitable for experimental observations of such phenomenon because of the existence of flattened regions of both the acoustic and exchange branches of the spin-wave spectrum.

In order to avoid any possible misunderstanding, we shall give the definition of the acoustic and exchange branches of the spin-wave spectrum of a many-sublattice magnet. The most convenient way to do it is to consider homogeneous oscillations corresponding to those branches called the modes of the magnetic resonance spectrum. The modes whose activation energies $\omega_\nu(0)$ [$\nu$ is the mode number, and $\omega_\nu(\mathbf{k})$ is the energy of the corresponding spin-wave branch] tend to zero in the exchange approximation will be referred to as the acoustic modes. There are three such modes in the case of a magnet noncollinear in the exchange approximation The precession of the magnetic moments of the sublattices taking part in the oscillations corresponding to the acoustic modes may be described in the following way. The magnetic structure formed by the spins of sublattices entering the magnetic unit cell rotates as a whole without inducing changes of the angles between the spins. The spin-wave branches corresponding to the acoustic modes are called acoustic spin waves. The rest of the $n-3$ modes of the resonance spectrum of the exchange-noncollinear $n$-sublattice magnet have the name "exchange modes." The activation energies of these branches are nonzero in the exchange approximation and are determined by different combinations of the inter- and intrasublattice exchange interaction parameters. The precession of the spins taking part in the oscillations corresponding to the given exchange mode leads to changes of the relative orientations of the spins inside the unit cell. The magnetic structure of the unit cell does not rotate as a whole in the case of oscillations corresponding to the exchange mode. In this respect exchange magnons are analogous to optical phonons because the spins of the sublattices entering the magnetic unit cell oscillate out of phase.

One has to distinguish between the terms "exchange" magnons and "optical" magnons. The last was used recently for the excitations which exist in magnetic crystals with $S \geq 1$ and strong single-ion anisotropy.[2] These exci-

 

tations correspond to transitions with nonconservation of the spin projection and their energies may be of the order of the exchange energy. The presence or absence of optical magnons is in no way connected with the number of magnetic sublattices in a magnet. They can, for example, exist even in a ferromagnet which has only one magnetic atom in the unit cell.

It is also necessary to note that in earlier papers in the 1960s and 1970s the exchange magnons under the above classification scheme were usually named optical magnons because of their analogy to optical phonons.

The order of magnitude of the absorption frequencies for acoustic spin-wave branches may be estimated to be $\omega \propto \sqrt{JA}$, where $J$ is the intersublattice exchange interaction parameter and $A$ is the anisotropy parameter, whereas two-magnon absorption by exchange branches takes place at the frequencies $\omega \propto J$. In the case of three-dimensional magnets, the flattening of the dispersion curves takes place only for exchange magnons because of the changes of the structure of the spectrum under the action of the external magnetic field.[3] It has to be remembered that in three-dimensional magnets two-magnon absorption by acoustic magnons at the frequencies $\omega \propto \sqrt{JA}$ may be observed only under the conditions of the so-called parametric excitation of spin waves.[4] It is caused by the small value of the density of the magnon states in this region of frequencies. The absorption intensity during the parametric excitation of the spin waves has the threshold character of the dependency on the intensity of the incident radiation (for details on the parametric excitation of the spin waves in antiferromagnets, see Ref. 5).

The coupling between the electric component of the electromagnetic radiation and the spin system of a magnetic crystal is stronger than that of the magnetic component. Thus, in crystals whose symmetry allows the existence of the so-called electric-dipole channel of the two-magnon absorption, the intensity of it will be much greater. One of the important advantages of the method of two-magnon absorption is that it allows one to study those branches of the spin-wave spectrum which cannot be observed in experiments on direct absorption (magnetic resonance) or on Raman light scattering. A similar situation takes place, for example, in $Nd_2CuO_4$. The exchange mode in this compound is determined by the product of the quadratic with respect to spin exchange interactions and four-site (biquadratic) one[23,13] (see Sec. II of the present paper). The four-site exchange interaction in turn is responsible for the stabilization of the plane-cross noncollinear magnetic ordering in this antiferromagnet. Because of the magnetic symmetry of $Nd_2CuO_4$ the exchange mode cannot be excited by external alternating magnetic field (see Ref. 23); that is, it cannot be observed by the magnetic resonance method.

As already noted, the intensity of two-magnon absorption is proportional to the density of magnon states. Thus the study of two-magnon absorption gives information about the density of the magnon states, which in turn allows one to make certain conclusions on the character of the critical points of the magnon spectrum.

The following note is also in order. Since the combined energy of the magnons excited during the two-magnon absorption process is fixed (equal to the frequency of the electromagnetic wave) in the experiments with a fixed frequency of the incident radiation and variable constant external magnetic field, one can study the dependences of the magnon frequencies on the wave vector.

By now, two-magnon absorption by acoustic branches of the spin wave spectrum has been investigated both theoretically and experimentally in considerable detail for quasi-two-dimensional two-sublattice antiferromagnets.[6–9] The peculiarities of two-magnon absorption by exchange spin waves have been considered in Refs. 10 and 11 and then in Ref. 3. Two-magnon absorption by exchange magnons in the regions of the spectrum corresponding to the flattened parts of the dispersion curves has been studied in these papers both theoretically and experimentally for the case of the three-dimensional four-sublattice antiferromagnet $CuCl_2 \cdot 2H_2O$.

The energy of exchange magnons in quasi-two-dimensional antiferromagnets at zero value of the wave vector, $\omega_E \propto \sqrt{JJ'}$, may be of the order of magnitude of the energy of acoustic magnons, $\omega_A \propto \sqrt{JA}$, because the inequality $J' \ll J$ takes place as a result of the quasi-two-dimensionality of the crystal. The theory of two-magnon absorption for this type of many-sublattice antiferromagnet has been considered in Ref. 12. In all these papers, the case of so-called many-sublattice magnets collinear in the exchange approximation has been employed. Two-magnon absorption in these antiferromagnets had a magnetic-dipole character. It means that the absorption of the electromagnetic wave in these crystals is due to the interaction of the magnetic component of the electromagnetic field with the crystal. However, two-magnon absorption in exchange-noncollinear magnets has not been

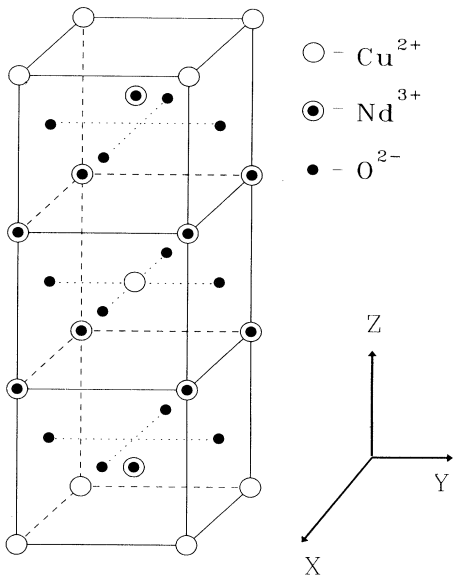


FIG. 1. Crystal structure of $Nd_2CuO_4$ in the paramagnetic state.

considered at all so far.

The exchange-noncollinear four-sublattice antiferromagnet $Nd_2CuO_4$ is unique from the point of view of experiment. First of all, the spin-wave spectrum of this antiferromagnet has a quasi-two-dimensional character for some particular directions of the wave vector.[13] Second, the special magnetic and crystal symmetry of this compound leads to the situation that the two-magnon absorption process is electric-dipole active in the exchange approximation.

The main goal of the present investigation is to analyze the peculiarities of two-magnon absorption in exchange-noncollinear many-sublattice antiferromagnets which symmetry allows the simultaneous existence of two channels of absorption due to electric-dipole and magnetic-dipole mechanisms.

In Sec. II a brief description of the spin-wave spectrum of the four-sublattice exchange-noncollinear antiferromagnet $Nd_2CuO_4$ will be given. Section III deals with the calculations of the frequency dependences of the coefficients of two-magnon absorption and the numerical analysis of the theoretical results obtained. Brief concluding remarks are given in the final section.

## II. SPIN-WAVE SPECTRUM

It is a common knowledge now[14-22] that $Nd_2CuO_4$ has tetragonal symmetry (the crystal structure is shown in Fig. 1) at temperatures above the Néel temperature $T_N=240$ K. Below the Néel temperature, it is the four-sublattice antiferromagnet with the magnetic structure of the "plane-cross" type in the absence of an external magnetic field. The sequence of magnetic phase transitions in the copper subsystem of this compound takes place during the lowering of the temperature. The ordering of neodymium subsystem occurs in the helium temperature region.[15,19,20] The theoretical analysis of the magnetically ordered phases possible in $Nd_2CuO_4$ and conditions of their stability in the absence of an external magnetic field has been carried out in Ref. 23. The behavior of the magnetic structure under the action of the external magnetic field has been studied in Ref. 24.

The general expression for the quadratic with respect to spins part of the Hamiltonian of the many-sublattice magnet may be written in the from

$$H=\sum_{i,j,\alpha,\beta} I^{ij}_{\alpha\beta}(m,n)S^{\alpha}_{i}(m)S^{\beta}_{j}(n) ,$$

where $I^{ij}_{\alpha\beta}(m,n)$ are the parameters of the sublattice interactions. The indices $\alpha,\beta$ label the sublattices; the indices $i,j$ are the Cartesian coordinates, and the indices $n$ and $m$ number the magnetic unit cells. Following Refs. 23 and 13, we introduce the linear combinations of the spins of the sublattices in Fourier representation:

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

We use here the same numbering scheme for the copper ions as in Ref. 23.

In terms of the vectors (1), the Hamiltonian of the copper magnetic subsystem of $Nd_2CuO_4$ may be represented as[13]

$$H=\sum_{\mathbf{k}}\{H^{(2)}(\mathbf{k})+H^{(4)}(\mathbf{k})\} , \tag{2}$$

where $H^{(2)}(\mathbf{k})$ and $H^{(4)}(\mathbf{k})$ describe the two- and four-site spin-spin interactions, respectively,

$$\begin{aligned}
H^{(2)}(\mathbf{k})=&J_{0j}(\mathbf{k})F_j(-\mathbf{k})F_j(\mathbf{k})+\sum_{\sigma=1}^{3}J_{\sigma j}(\mathbf{k})L_{\sigma j}(-\mathbf{k})L_{\sigma j}(\mathbf{k})+d_1(\mathbf{k})L_{1x}(-\mathbf{k})L_{2y}(\mathbf{k})+d_2(\mathbf{k})L_{1y}(-\mathbf{k})L_{2x}(\mathbf{k})\\
&+d_3(\mathbf{k})F_x(-\mathbf{k})L_{3y}(\mathbf{k})+d_4(\mathbf{k})F_y(-\mathbf{k})L_{3x}(\mathbf{k}) .
\end{aligned} \tag{3}$$

The $J_{\sigma j}(\mathbf{k})$ and $d(\mathbf{k})$ values in expression (3) are the parameters of the so-called two-site exchange interactions and Dzyaloshinsky-Moriya interactions, respectively.[23] The $z$ axis of the coordinate system is chosen to be parallel to the tetragonal crystal axis. In this case the $x,y$ planes coincide with the $CuO_2$ planes of the crystal lattice.

In the nearest-neighbor approximation, the following expressions for $J_{\sigma j}(\mathbf{k})$ may be obtained[13]

$$\begin{aligned}
&J_{0j}(\mathbf{k})=I^{jj}_{11}\gamma_1(\mathbf{k})+I^{jj}_{12}\gamma_2(\mathbf{k})+I^{jj}_{13}\gamma_3(\mathbf{k})+I^{jj}_{14}\gamma_4(\mathbf{k}) ,\\
&J_{1j}(\mathbf{k})=I^{jj}_{11}\gamma_1(\mathbf{k})+I^{jj}_{12}\gamma_2(\mathbf{k})-I^{jj}_{13}\gamma_3(\mathbf{k})-I^{jj}_{14}\gamma_4(\mathbf{k}) ,\\
&J_{2j}(\mathbf{k})=I^{jj}_{11}\gamma_1(\mathbf{k})-I^{jj}_{12}\gamma_2(\mathbf{k})+I^{jj}_{13}\gamma_3(\mathbf{k})-I^{jj}_{14}\gamma_4(\mathbf{k}) ,\\
&J_{3j}(\mathbf{k})=I^{jj}_{11}\gamma_1(\mathbf{k})-I^{jj}_{12}\gamma_2(\mathbf{k})-I^{jj}_{13}\gamma_3(\mathbf{k})+I^{jj}_{14}\gamma_4(\mathbf{k}) .
\end{aligned} \tag{4}$$

Here

$$\begin{aligned}
&\gamma_1(\mathbf{k})=\cos\frac{\mathbf{k}\cdot(\hat{\mathbf{a}}+\hat{\mathbf{b}})}{2}\cos\frac{\mathbf{k}\cdot(\hat{\mathbf{a}}-\hat{\mathbf{b}})}{2} ,\\
&\gamma_2(\mathbf{k})=\cos\frac{\mathbf{k}\cdot\hat{\mathbf{a}}}{2}\cos\frac{\mathbf{k}\cdot\hat{\mathbf{c}}}{2}\cos 2(\mathbf{k}\cdot\Delta_y) ,\\
&\gamma_3(\mathbf{k})=\cos\frac{\mathbf{k}\cdot\hat{\mathbf{b}}}{2}\cos\frac{\mathbf{k}\cdot\hat{\mathbf{c}}}{2}\cos 2(\mathbf{k}\cdot\Delta_x) ,\\
&\gamma_4(\mathbf{k})=\cos\frac{\mathbf{k}\cdot\hat{\mathbf{a}}}{2}\cos\frac{\mathbf{k}\cdot\hat{\mathbf{b}}}{2} ,
\end{aligned}$$

where $\hat{\mathbf{a}}$, $\hat{\mathbf{b}}$, and $\hat{\mathbf{c}}$ are the lattice vectors directed along the $x$, $y$, and $z$ axes, respectively; the $\Delta_x,\Delta_y$ are the lattice distortions which appear as a result of the structural phase transition[14] above $T_N$. The role of the structural

distortions in the formation of the magnetic ordering of $Nd_2CuO_4$ has been discussed in Ref. 23. The coefficients $d_\alpha(\mathbf{k})$ may be represented by the expressions[13]

$$\begin{aligned} d_1(\mathbf{k})&=2[I_{11}^{xy}\gamma_1(\mathbf{k})-I_{14}^{xy}\gamma_4(\mathbf{k})-2I_{12}^{xy}\gamma_5(\mathbf{k})]\ , \\ d_2(\mathbf{k})&=2[I_{11}^{xy}\gamma_1(\mathbf{k})-I_{14}^{xy}\gamma_4(\mathbf{k})+2I_{12}^{xy}\gamma_5(\mathbf{k})]\ , \\ d_3(\mathbf{k})&=2[I_{11}^{xy}\gamma_1(\mathbf{k})+I_{14}^{xy}\gamma_4(\mathbf{k})-2I_{12}^{xy}\gamma_6(\mathbf{k})]\ , \\ d_4(\mathbf{k})&=2[I_{11}^{xy}\gamma_1(\mathbf{k})+I_{14}^{xy}\gamma_4(\mathbf{k})+2I_{12}^{xy}\gamma_6(\mathbf{k})]\ , \end{aligned} \tag{5}$$

in which

$$\gamma_5(\mathbf{k})=\cos\frac{\mathbf{k}\cdot\hat{\mathbf{c}}}{2}\cos\frac{\mathbf{k}\cdot(\hat{\mathbf{a}}+\hat{\mathbf{b}})}{4}\cos\frac{\mathbf{k}\cdot(\hat{\mathbf{a}}-\hat{\mathbf{b}})}{4}\ ,$$

$$\gamma_6(\mathbf{k})=\cos\frac{\mathbf{k}\cdot\hat{\mathbf{c}}}{2}\sin\frac{\mathbf{k}\cdot(\hat{\mathbf{a}}+\hat{\mathbf{b}})}{4}\sin\frac{\mathbf{k}\cdot(\hat{\mathbf{a}}-\hat{\mathbf{b}})}{4}\ .$$

Later on, we will neglect the small values $\Delta_i$ in comparison with the lattice parameters where appropriate.

Considering the four-site exchange interaction, we restrict ourselves to the interaction of the two nearest pairs of spins of the copper ions from the adjacent $x,y$ layers. The expression for $H^{(4)}(\mathbf{k})$ in this approximation may be written in the form[13]

$$\begin{aligned} H^{(4)}(\mathbf{k})=\frac{1}{N}\sum_{\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} D\{&[\mathbf{S}_1(\mathbf{k})\cdot\mathbf{S}_2(\mathbf{k}_2)][\mathbf{S}_3(\mathbf{k}_3)\cdot\mathbf{S}_4(\mathbf{k}_4)] \\ &+[\mathbf{S}_1(\mathbf{k})\cdot\mathbf{S}_3(\mathbf{k}_3)][\mathbf{S}_2(\mathbf{k}_2)\cdot\mathbf{S}_4(\mathbf{k}_4)]\} \\ &\times\Phi(\mathbf{k},\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4)\ , \end{aligned} \tag{6}$$

where

$$\Phi(\mathbf{k},\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4)=\cos\frac{\hat{\mathbf{a}}\cdot\mathbf{K}_2}{4}\cos\frac{\hat{\mathbf{b}}\cdot\mathbf{K}_1}{4}\cos\frac{\hat{\mathbf{c}}\cdot\mathbf{K}_3}{4}\Delta(\mathbf{K}_0)\ ,$$

and

$$\mathbf{K}_0=\mathbf{k}+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4,\quad \mathbf{K}_1=\mathbf{k}+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4\ ,$$

$$\mathbf{K}_2=\mathbf{k}-\mathbf{k}_2+\mathbf{k}_3-\mathbf{k}_4,\quad \mathbf{K}_3=\mathbf{k}-\mathbf{k}_2-\mathbf{k}_3+\mathbf{k}_4\ .$$

$N$ is the number of the unit cells of the crystal, and $\Delta(\mathbf{k})$ is the Kronecker symbol.

A comprehensive analysis of the spin-wave spectrum of $Nd_2CuO_4$ in the noncollinear magnetic phase (see Fig. 2) which corresponds to the $\tau_2$ irreducible representation of the $D_{4h}^{14}$ symmetry group has been carried out in Ref. 13. We will present here the main results of this analysis. As has been revealed by neutron experiments,[14,17,18,21,22] the noncollinear magnetic phase of $Nd_2CuO_4$ has been realized in the temperature intervals 4 K $<T<$ 30 K and 70 K $<T<T_N$. The magnetic structure of this phase is formed by two antiferromagnetically ordered $x$-$y$ planes of copper ions (see Figs. 1 and 2). The nonzero equilibrium values of the antiferromagnetic vectors of these planes are equal to $\bar{L}_{1x}=\bar{L}_{2y}=4\sqrt{2}S$ in the absence of an external field.

The quantization of the Hamiltonian (2) along with (3) and (6) was carried out with the help of the approach developed in Ref. 25. The part of the Hamiltonian (2) quadratic with respect to boson spin-deflection operators

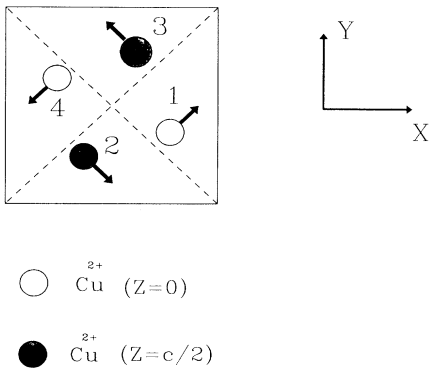


FIG. 2. Magnetic unit cell for the noncollinear magnetic phase. Solid and open circles denote the copper ions located in the $c/2$ distant crystal planes.

has the form[13]

$$H=\frac{1}{2}\sum_{L,\mathbf{k}}\{q_L(\mathbf{k})Q_L(\mathbf{k})Q_L(-\mathbf{k})-p_L(\mathbf{k})P_L(\mathbf{k})P_L(-\mathbf{k})\}\ . \tag{7}$$

In this expression,

$$Q_L(\mathbf{k})=\frac{1}{\sqrt{2}}[a_L^\dagger(-\mathbf{k})+a_L(\mathbf{k})]\ ,$$

$$P_L(\mathbf{k})=\frac{1}{\sqrt{2}}[a_L^\dagger(\mathbf{k})-a_L(-\mathbf{k})]\ ,$$

where $a_L^\dagger(\mathbf{k}),a_L(\mathbf{k})$ are the linear combinations of boson spin-deflection operators of the sublattices, $L=L_0$, $L_1$, $L_2$, and $L_3$, and $L_0=F$. The coefficients $q_L(\mathbf{k})$ and $p_L(\mathbf{k})$ from (7) are determined by the expressions

$$p_L(\mathbf{k})=2E_{L,z}(\mathbf{k})\ , \tag{8}$$

$$\begin{aligned} q_{L_0}(\mathbf{k})&=E_{2,x}(\mathbf{k})+E_{1,y}(\mathbf{k})-4Sd_2(\mathbf{k}) \\ &\quad+8S^3D[2\gamma_4(\mathbf{k})-\gamma_2(\mathbf{k})-\gamma_3(\mathbf{k})]\ , \\ q_{L_1}(\mathbf{k})&=E_{0,y}(\mathbf{k})+E_{3,x}(\mathbf{k})-4Sd_4(\mathbf{k}) \\ &\quad+8S^3D[2\gamma_4(\mathbf{k})-\gamma_2(\mathbf{k})+\gamma_3(\mathbf{k})]\ , \\ q_{L_2}(\mathbf{k})&=E_{0,x}(\mathbf{k})+E_{3,y}(\mathbf{k})-4Sd_3(\mathbf{k}) \\ &\quad+8S^3D[2\gamma_4(\mathbf{k})+\gamma_2(\mathbf{k})-\gamma_3(\mathbf{k})]\ , \\ q_{L_3}(\mathbf{k})&=E_{1,x}(\mathbf{k})+E_{2,y}(\mathbf{k})-4Sd_1(\mathbf{k}) \\ &\quad+8S^3D[2\gamma_4(\mathbf{k})+\gamma_2(\mathbf{k})+\gamma_3(\mathbf{k})]\ , \end{aligned} \tag{9}$$

and

$$E_{L_{\sigma,j}}=4S[J_{\sigma j}(\mathbf{k})-J_{1x}(0)-\tfrac{1}{2}d_1(0)],\quad j=x,y,z\ . \tag{10}$$

The standard Bogolyubov transformation of the $a_L(\mathbf{k})$ operators into $\xi_\nu^\dagger(\mathbf{k})$ and $\xi_\nu(\mathbf{k})$ operators of creation and annihilation of a magnon of the $\nu$th branch of the spectrum,

$$Q_L(\mathbf{k})=\frac{1}{\sqrt{2}}t_{L\nu}(\mathbf{k})[\xi^{\dagger}_{\nu}(-\mathbf{k})+\xi_{\nu}(\mathbf{k})] ,$$

$$P_L(\mathbf{k})=\frac{1}{\sqrt{2}}d_{L\nu}(\mathbf{k})[\xi^{\dagger}_{\nu}(\mathbf{k})-\xi_{\nu}(-\mathbf{k})] ,$$

has been used. The $t_{L\nu}(\mathbf{k})$ and $d_{L\nu}(\mathbf{k})$ coefficients are determined by

$$\begin{aligned} t_{L\nu}(\mathbf{k})&=[p_L(\mathbf{k})/q_L(\mathbf{k})]^{1/4} , \\ d_{L\nu}(\mathbf{k})&=[q_L(\mathbf{k})/p_L(\mathbf{k})]^{1/4} . \end{aligned} \tag{11}$$

The following expressions for the energies of the spin waves have been obtained:[13]

$$\begin{aligned} \omega_{A_1}(\mathbf{k})&=\sqrt{p_{L_1}(\mathbf{k})q_{L_1}(\mathbf{k})} , \\ \omega_{A_2}(\mathbf{k})&=\sqrt{p_{L_2}(\mathbf{k})q_{L_2}(\mathbf{k})} , \\ \omega_{A_3}(\mathbf{k})&=\sqrt{p_{L_0}(\mathbf{k})q_{L_0}(\mathbf{k})} , \\ \omega_E(\mathbf{k})&=\sqrt{p_{L_3}(\mathbf{k})q_{L_3}(\mathbf{k})} . \end{aligned} \tag{12}$$

At $\mathbf{k}=0$ we get four modes of magnetic resonance with $\nu=A_1, A_2, A_3$ and $E$ as obtained in Ref. 23. The energies of the two acoustic modes $A_1$ and $A_2$ are degenerate.

Let us briefly discuss the dependences of the spin-wave spectrum branches on the wave vector for different orientations. In the case $\mathbf{k}\|[001]$, the energies of the modes $A_1$ and $A_2$ are degenerate and do not depend on the wave vector. This particular case of the absence of the dispersion dependences of the $A_1$ and $A_2$ branches is connected with the specific crystal and magnetic symmetry of $Nd_2CuO_4$ (see Ref. 13 for details). Thus in the case $\mathbf{k}\|[001]$ the spin-wave modes $A_1$ and $A_2$ manifest a pure two-dimensional behavior. As to the exchange $E$ mode, it has a very weak dispersion and its energy is approximately equal to the value corresponding to $\omega_E(0)$. That is, the dispersion relation of this mode also has a quasi-two-dimensional character.

In the case when the wave vector is oriented in such a way that it has components in the plane parallel to the $CuO_2$ layers (the $x$-$y$ plane in our case), the dispersion relations of the spin-wave branches are typical of a three-dimensional magnet. Similar to all the parent compounds of the high-temperature superconductors, the intralayer exchange interaction is much greater than the others. As a result, the spin-wave frequencies have a linear dependence on the wave vector in a wide enough region of the wave vectors, $\omega_0\omega_{\mathrm{Br}}^{-1}\ll(ak)\ll 1$, where $\omega_0$ is the value of the frequency at $\mathbf{k}=0$ and $\omega_{\mathrm{Br}}$ is that at the edge of the Brillouin zone.

The values of the parameters which describe spin-spin interactions in the copper magnetic subsystem of $Nd_2CuO_4$ [see expressions (3) and (6)] may be roughly estimated on the basis of the experimental results of Refs. 26–28 (see also Ref. 13). These parameters allow us to carry out the illustrative numerical calculations of the dependences of the spin-wave frequencies on the wave vector. The frequencies of the homogeneous vibrations corresponding to the acoustic branches of the magnon spectrum of $Nd_2CuO_4$ may be represented as[23,13]

$$\begin{aligned} \omega_{A_1}(0)&=\omega_{A_2}(0)=16S\sqrt{-Ja_2} , \\ \omega_{A_3}(0)&=16S\sqrt{-J(a_2-a_4)} , \end{aligned} \tag{13}$$

where $J\simeq I_{14}$ is the combination of the exchange parameters, and $a_i$ $(i=2,4,6,8)$ are the anisotropy constants (see Refs. 23 and 13 for details). The value of the intralayer exchange interaction $I_{14}=870\ \mathrm{cm}^{-1}$ was obtained in Ref. 26. As already noted, the interlayer exchange interaction is supposedly the largest in the given crystal. Using the experimental results[27,28] and the formulas (13), we estimated the values of $a_2$ and $(a_2-a_4)$ and obtained[13]

$$|a_2|\simeq 2.87\times 10^{-4}\ \mathrm{cm}^{-1},\quad |a_2-a_4|\simeq 3.32\times 10^{-5}\ \mathrm{cm}^{-1} .$$

Other anisotropy constants have to be of the same order of magnitude, because each of them contains the "large" intralayer anisotropy.[13] The differences $a_2-a_4$ and $a_6-a_8$ in turn are determined by the weaker interlayer anisotropy. The values of the parameters of the four-site $D$ and $I_{12}$ of the interlayer exchange interactions are unknown at present. The results of the estimations allowed us to suppose that the inequality

$$J\gg D\gg A \tag{14}$$

is satisfied, where $A$ is the combinations of the anisotropy constants.

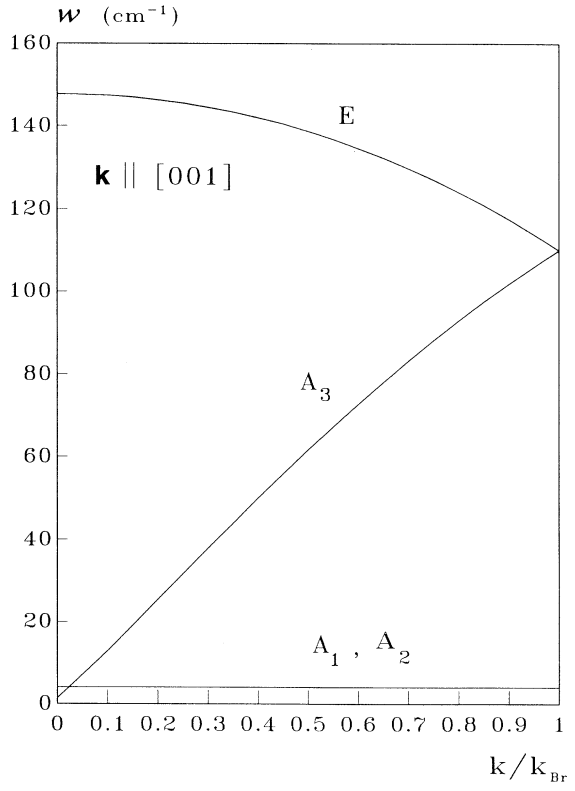


FIG. 3. Spin-wave spectrum for the wave vector oriented along the tetragonal crystal axis $\mathbf{k}\|[001]$.

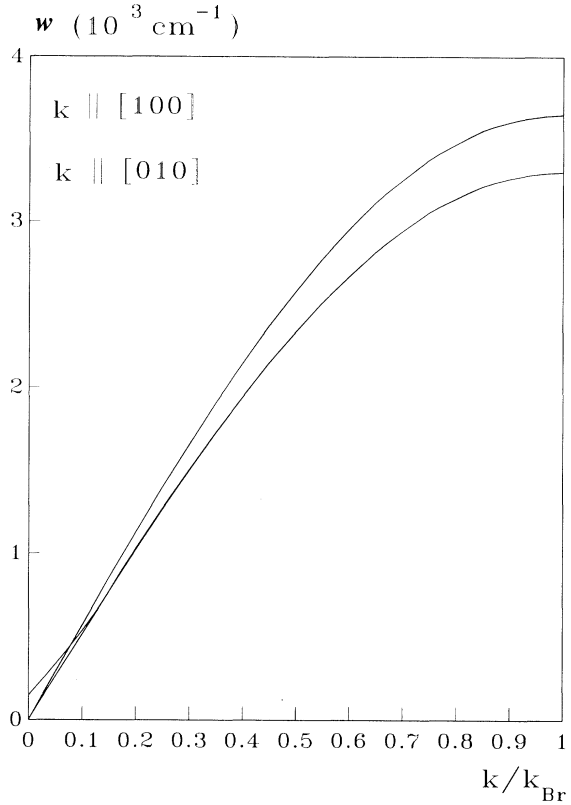


FIG. 4. Dependences of the spin-wave energies on the wave vector oriented along the [100] or [010] direction.

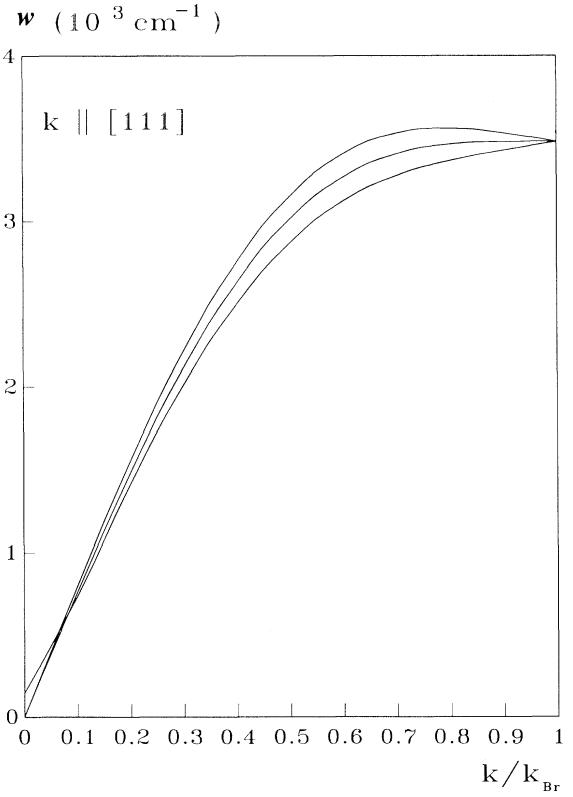


FIG. 5. Dispersion relations of the spin waves in the case of the orientation of the wave vector along the [111] direction.

We give here the results of the calculations of the spin-wave spectrum of $Nd_2CuO_4$ for the values of $I_{12}$ and $D$ related to the known value of the exchange parameter $I_{14}$ as follows:

$$I_{12}=10^{-1}I_{14}, \quad D=10^{-3}I_{14} . \tag{15}$$

During the calculations, we have neglected the value $I_{11}$ in comparison with $I_{14}$.

The dependences of the energies of the acoustic and exchange spin waves on the wave vector oriented along the direction [001] are shown in Fig. 3. The directions of the wave vector, [100] and [010], are equivalent, and the corresponding spectrum is given in Fig. 4. The dependences of the spin-wave frequencies on the wave vector along the [111] direction are depicted in Fig. 5.

## III. TWO-MAGNON ABSORPTION OF ELECTROMAGNETIC WAVES

The interaction Hamiltonian of the magnetic system with a uniform external electromagnetic field may be written as

$$H_{\rm int}=\sqrt{N}\{-g_{ij}\mu_B h_i F_j(0)+e_j P_j(0)\} , \tag{16}$$

where $h_i$ and $e_i$ are the components of the magnetic and electric vectors of the electromagnetic wave, $i,j=x,y,z$; $g_{ij}$ is the $g$-factor tensor; $F_i(0)$ is the zero Fourier component of the magnetization vector of the many-sublattice magnet [see formulas (1)]; and $P_i(0)$ is the zero Fourier component of the electric-dipole moment associated with the pair of spins.[29] According to Ref. 29, the general expression for the electric-dipole moment associated with the pair of spins of a magnetic crystal may be represented as

$$P_{lm}^{j}=\pi_{lm}^{j}(\mathbf{S}_l\cdot\mathbf{S}_m)+\sum_{r,p}\Gamma_{lm}^{j,rp}(S_{lr}S_{mp}+S_{mr}S_{lp}) + \sum_{r} d_{lm}^{j,r}[\mathbf{S}_l\times\mathbf{S}_m]_r .$$

In this expression the indices $r$ and $p$ are Cartesian coordinates; the indices $l$ and $m$ number the positions of the ions. Possible forms of the tensors $\pi_{lm}$, $\Gamma_{lm}^{j,rp}$, and $d_{lm}^{j,r}$ are determined by the positional symmetry of the ions and the pair of the ions $l$ and $m$. Several microscopic mechanisms can give rise to the electric-dipole moment associated with the pair of spins.[29] These mechanisms have been discussed in Ref. 29 where one can also find references on the original papers where they have been proposed. These mechanisms are the interionic Coulomb interaction of the exchange type and multipole-multipole type, the spin-orbit interaction, and some others.[29] Magnetic symmetry of $Nd_2CuO_4$ permits the existence of the exchange contribution into the electric-dipole moment associated with the pair of spins. With respect to Ref. 29 (see also Ref. 30 where this mechanism was considered), it is given by the second-order perturbation including nondiagonal exchange energy and electric-dipole moment. The contribution of this mechanism is described by the first term in the above expression. In what follows

we are not going to consider any other mechanisms which contribute to the vector **P**.

Thus the electric-dipole moment associated with the pair of spins in our case may be written in the form

$$P_j(0)=\sum_{\mathbf{k}} \pi^j_{\alpha\beta}(\mathbf{k})[\mathbf{S}_\alpha(-\mathbf{k})\cdot\mathbf{S}_\beta(\mathbf{k})] \ . \tag{17}$$

The Greek indices numerate the magnetic sublattices in this expression. The tensors $\pi^j_{\alpha\beta}(0)$ are nonzero for those pairs of copper ions which cannot be permuted by the inversion operation and also for ions the positional symmetry of which does not contain an inversion. That is why for the crystal symmetry under consideration the tensor $\pi^j_{\alpha\beta}(0)$ is equal to zero in the case where the structural distortions $\Delta_i$ are absent.

Using relations (1) and the structure of the tensors $\pi^j_{\alpha\beta}$ as discussed above, one can get the following expression for the components of the polarization vector (17) in the nearest-neighbor approximation:

$$\begin{aligned} P_x(0)=&\frac{1}{2}\sum_{\mathbf{k}}\{[\pi_{11}\gamma_1(\mathbf{k})+\pi_{12}\gamma_2(\mathbf{k})]F(-\mathbf{k})L_1(\mathbf{k}) \\ &+[\pi_{11}\gamma_1(\mathbf{k})-\pi_{12}\gamma_2(\mathbf{k})]L_2(-\mathbf{k})L_3(\mathbf{k})\} \ , \\ P_y(0)=&\frac{1}{2}\sum_{\mathbf{k}}\{[\pi_{11}\gamma_1(\mathbf{k})+\pi_{12}\gamma_2(\mathbf{k})]F(-\mathbf{k})L_2(\mathbf{k}) \\ &+[\pi_{11}\gamma_1(\mathbf{k})-\pi_{12}\gamma_2(\mathbf{k})]L_1(-\mathbf{k})L_3(\mathbf{k})\} \ . \end{aligned} \tag{18}$$

The $P_z(0)$ component of the electric-dipole moment associated with the pair of spins is equal to zero in the exchange approximation. Note that in obtaining (18) we have utilized the relations

$$\pi^x_{11}(0)=\pi^y_{11}(0)=\pi_{11}, \quad \pi^x_{12}(0)=\pi^y_{12}(0)=\pi_{12} \ .$$

Quantization of the Hamiltonian (16) along with (17) and (18) has been carried out with the help of the same quantization procedure of the Hamiltonian (2). The quadratic with respect to the creation and annihilation operators of the spin-wave part of expression (16) maybe represented as the sum of two contributions:

$$\begin{aligned} H^{(2)}_{\text{int}}&=H^{(2,m)}_{\text{int}}+H^{(2,e)}_{\text{int}} \\ &=\sum_{\mathbf{k}}\{[f^{(h)}_{\mu\nu}(\mathbf{k})+f^{(e)}_{\mu\nu}(\mathbf{k})]\xi^\dagger_\mu(\mathbf{k})\xi^\dagger_\nu(-\mathbf{k})+\text{H.c.}\} \ . \end{aligned} \tag{19}$$

The Hamiltonian $H^{(2,m)}_{\text{int}}$ describes the magnetic-dipole channel of two-magnon absorption,

$$\begin{aligned} H^{(2,m)}_{\text{int}}=&\frac{g_{xx}\mu_B h_x}{2\sqrt{2}}\sum_{\mathbf{k}}\{\Phi(q_0,p_0;q_1,p_1)\xi^\dagger_{A_1}(\mathbf{k})\xi^\dagger_{A_3}(-\mathbf{k}) \\ &+\Phi(q_2,p_2;q_3,p_3)\xi^\dagger_{A_2}(\mathbf{k})\xi^\dagger_E(-\mathbf{k})+\text{H.c.}\} \\ &+[x\Longrightarrow y; \ \ q_1,p_1\Longleftrightarrow q_2,p_2; \ \ A_2\Longleftrightarrow A_1] \ , \end{aligned} \tag{20}$$

where

$$\Phi(q_\alpha,p_\alpha,q_\beta,p_\beta)=(q_\alpha q_\beta/p_\alpha p_\beta)^{1/4}-(p_\alpha p_\beta/q_\alpha q_\beta)^{1/4} \ .$$

The expressions for $p_\sigma=p_\sigma(\mathbf{k})$ and $q_\sigma=q_\sigma(\mathbf{k})$ are given in the previous section by formulas (8)–(10) where $\sigma=0,1,2,3$ correspond to $L=F,L_1,L_2,L_3$. The Hamiltonian $H^{(2,e)}_{\text{int}}$ represents the electric-dipole channel of two-magnon absorption,

$$\begin{aligned} H^{(2,e)}_{\text{int}}=&-e^x S\sum_{\mathbf{k}}\{\Psi^{(-)}(q_0,p_0;q_1,p_1)\xi^\dagger_{A_1}(\mathbf{k})\xi^\dagger_{A_3}(-\mathbf{k}) \\ &+\Psi^{(+)}(q_2,p_2;q_3,p_3)\xi^\dagger_{A_2}(\mathbf{k})\xi^\dagger_E(-\mathbf{k})+\text{H.c.}\} \\ &+[x\Longrightarrow y; \ \ q_1,p_1\Longleftrightarrow q_2,p_2; \ A_2\Longleftrightarrow A_1;\gamma_2\Longleftrightarrow\gamma_1] \ , \end{aligned} \tag{21}$$

where

$$\begin{aligned} \Psi^{(\pm)}(q_\alpha,p_\alpha;q_\beta,p_\beta)=&\pi_{11}[1-\gamma_1(\mathbf{k})][(q_\alpha q_\beta/p_\alpha p_\beta)^{1/4} \\ &-(p_\alpha p_\beta/q_\alpha q_\beta)^{1/4}] \\ &\pm\pi_{12}\gamma_2(\mathbf{k})(q_\alpha q_\beta/p_\alpha p_\beta)^{1/4} \ . \end{aligned}$$

Two-magnon absorption does not take place when the magnetic component of the electromagnetic wave is oriented along the $z$ axis. To obtain this contribution in the interaction Hamiltonian for this geometry, one has to take into account three-magnon processes at second order of perturbation theory.[12] It follows from expressions (20), (21) that the processes of the photon decay have the same types for both the magnetic and electric-dipole channels. The exchange magnon participating in the absorption process is always accompanied by one acoustic spin wave which corresponds to the two-dimensional irreducible representation at $\mathbf{k}=0$. In contrast to three-dimensional collinear magnets,[3] the amplitude describing the two-magnon absorption process with the participation of the exchange magnon does not contain any small parameter. The absorption of the electromagnetic wave leading to the generation of magnons corresponding to the same branches of the spin-wave spectrum is absent.

The separation of the electric- and magnetic-dipole channels of the absorption may be carried out with the help of the proper choice of the orientation of the polarizations of the electromagnetic field components and the direction of the propagation of the incident radiation. For example, in the case when $\mathbf{k}\|oy$ and $\mathbf{e}\|oz$ only magnetic-dipole absorption by magnons of the $A_1, A_3$ and $E, A_2$ branches takes place. The absorption coefficient at low temperatures is described by the expression

$$\begin{aligned} K^{(h)}_{\mu,\nu}(\omega)=&\text{const}\times\omega\int d^3k\,|f^{(h_x)}_{\mu,\nu}(\mathbf{k})|^2 \\ &\times\delta(\omega-\omega_\mu(\mathbf{k})-\omega_\nu(\mathbf{k})) \ , \end{aligned} \tag{22}$$

in which $\mu,\nu=(A_1,A_3;E,A_2)$. For the same direction of the propagation of the incident radiation, but with the orientation of the electric component $\mathbf{e}\|ox$, two-magnon absorption by the branches $A_1, A_3$ and $E, A_2$ will be due to the electric-dipole mechanism only:

$$K^{(e)}_{\mu,\nu}(\omega)=\text{const}\times\omega\int d^3k\,|f^{(e_x)}_{\mu,\nu}(\mathbf{k})|^2 \times\delta(\omega-\omega_\mu(\mathbf{k})-\omega_\nu(\mathbf{k}))\ . \tag{23}$$

The structure of the amplitudes $f^{(h)}_{\mu\nu}(\mathbf{k})$ and $f^{(e)}_{\mu\nu}(\mathbf{k})$ may be obtained from expressions (19)–(21). During the calculations of the absorption coefficients (22) and (23), the interlayer exchange parameter $I_{12}$ has been neglected in comparison with the intralayer exchange $I_{14}$ where appropriate for simplicity's sake. This approximation does not change the qualitative behavior of the absorption coefficients $K_{\mu\nu}(\omega)$.

For magnetic-dipole absorption by the $A_1$ and $A_3$ branches in the frequency region $\omega\ll\omega_{\text{ex}}$, where $\omega_{\text{ex}}=8SI_{14}$, one can get

$$K^{(h)}_{A_1,A_3}(\omega)=\text{const}\times\frac{(g\mu_B)^2}{4\omega^2_{\text{ex}}\omega^2}\times\int^1_{t_0(\omega)}\frac{[\omega^2_{a_4}\text{sgn}(a_4)+\omega^2_E(1-t)]^2}{\sqrt{1-t^2}}dt\ . \tag{24}$$

Here we introduced the notation $\omega_{a_4}=16S\sqrt{I_{14}|a_4|}$; the frequency $\omega_E=16S\sqrt{I_{14}S^2D}$ is slightly different from the energy gap of the exchange spin-wave branch, which is equal to $\omega_E(0)=16S\sqrt{2(I_{14}-I_{12})S^2D}$. Also, we use the notation $\omega_{A_1}$, $\omega_{A_2}$, and $\omega_{A_3}$ for the energy gaps of the acoustic spin-wave branches which are determined by formulas (13).

The electric-dipole contribution to the absorption by $A_1$ and $A_3$ in the same region of the frequencies, $\omega\ll\omega_{\text{ex}}$, is given by the expression

$$K^{(e)}_{A_1,A_3}(\omega)=\text{const}\times\frac{(S\pi_{12})^2}{16\omega^2_{\text{ex}}\omega^2}\int^1_{t_0(\omega)}\frac{[\omega^2+\omega^2_{a_4}\text{sgn}(a_4)+\omega^2_E(1-t)]^2t}{\sqrt{1-t^2}}dt\ . \tag{25}$$

The limiting value $t_0(\omega)$ in formulas (24) and (25) is determined by the expressions

$$t_0(\omega)=\begin{cases}(\omega^2_E-\omega^2+\omega^2_{a_4}\text{sgn}(a_4)+2\omega\omega_{A_1})\omega_E^{-2} & \text{at } \omega_0\le\omega\le\omega_1,\\ 0 \text{ at } \omega_1\le\omega\ ,\end{cases} \tag{26}$$

in which

$$\omega_0=\omega_{A_3}+\omega_{A_1},\quad \omega_1=\omega_{A_1}+\sqrt{\omega^2_E+\omega^2_{A_3}}\ . \tag{27}$$

The coefficients of the magnetic- and electric-dipole absorption by the $A_2$ and $E$ modes may be represented as

$$K^{(h)}_{A_2,E}(\omega)=\text{const}\times\frac{(g\mu_B)^2}{4\omega^2_{\text{ex}}\omega^2}\int_0^{t_0(\omega)}\frac{[\omega^2_{a_6}\text{sgn}(a_6)+\omega^2_E(1+t)]^2}{\sqrt{1-t^2}}dt \tag{28}$$

and

$$K^{(e)}_{A_2,E}(\omega)=\text{const}\times\frac{(S\pi_{12})^2}{16\omega^2_{\text{ex}}\omega^2}\int_0^{t_0(\omega)}\frac{[\omega^2+\omega^2_{a_6}\text{sgn}(a_6)+\omega^2_E(1+t)]^2t}{\sqrt{1-t^2}}dt\ . \tag{29}$$

The limiting value $t_0(\omega)$ in these formulas is determined by

$$t_0(\omega)=\begin{cases}(\omega^2-\omega^2_{a_6}-\omega^2_E-2\omega\omega_{A_2})\omega_E^{-2} & \text{at } \omega_0\le\omega\le\omega_1,\\ 1 \text{ at } \omega_1\le\omega\ ,\end{cases} \tag{30}$$

and

$$\omega_0=\omega_{A_2}+\sqrt{\omega^2_E+\omega^2_{62}\text{sgn}(a_6-a_2)},\quad \omega_1=\omega_{A_2}+\sqrt{2\omega^2_E+\omega^2_{62}\text{sgn}(a_6-a_2)}\ , \tag{31}$$

where $\omega_{62}=16S\sqrt{I_{14}|a_6-a_2|}$. The calculations of the electric-dipole absorption in the region $\omega\ll\omega_{\text{ex}}$ was carried out without taking into account the contributions to $f^{(e)}_{A_1,A_3}$ proportional to $\pi_{11}[1-\gamma_1(\mathbf{k})]$. The omitted term contains the factor $1-\gamma_1(\mathbf{k})$, which is of the order of $\frac{1}{4}[(\mathbf{k}\cdot\hat{\mathbf{a}})^2+(\mathbf{k}\cdot\hat{\mathbf{b}})^2]\ll1$ in the region of low frequencies, $\omega\propto\sqrt{JA}$.

Following from formulas (24)–(27) and (28)–(31), the absorption coefficient due to the electric-dipole channel increases with increasing frequency, whereas the frequen-

cy dependence of the coefficient of magnetic-dipole absorption is nonmonotonic. Such a behavior of the absorption coefficient is connected with the specific features of the spin-wave spectrum and the structure of the amplitude $f^{(h)}_{\mu\nu}(\mathbf{k})$ describing the magnetic-dipole part of the interaction of the electromagnetic wave with the crystal. The quasi-two-dimensionality of the spin-wave spectrum of $Nd_2CuO_4$ leads to different behavior of the appropriate $t_{Lv}(\mathbf{k})$ and $d_{Lv}(\mathbf{k})$ coefficients of the unitary transformation in the two regions of the wave vector $\mathbf{k}$ space. In the first region determined by the inequality $(\mathbf{k}\cdot\hat{\mathbf{a}}),(\mathbf{k},\hat{\mathbf{b}}) \ll \sqrt{(D+a)I_{14}^{-1}}$, the $t_{Lv}(\mathbf{k})$ and $d_{Lv}(\mathbf{k})$ coefficients (11) do not depend or depend weakly on the $k_z$ component of the wave vector, which varies from 0 to $\pi/c$. So these coefficients are approximately equal to $t_{Lv}(0)$ and $d_{Lv}(0)$ in this region of wave vectors. The second region of the wave vector $\mathbf{k}$ space is determined by the inequality $\sqrt{(D+a)I_{14}^{-1}} \ll (\mathbf{k}\cdot\hat{\mathbf{a}}),(\mathbf{k}\cdot\hat{\mathbf{b}}) \ll 1$. The $t_{Lv}(\mathbf{k})$ and $d_{Lv}(\mathbf{k})$ coefficients each tends to unity in this region, and as a result, the $f^{(h)}(\mathbf{k})$ amplitude is proportional to the inverse square of the frequency. This nonmonotonic behavior of the dependence of the magnetic-dipole absorption coefficient on frequency will still retain even if one takes into account the interlayer exchange interaction coefficient $I_{12}$ during the calculations.

The frequency dependences of the absorption coefficients $K^{(h)}_{A_1,A_3}(\omega)$ and $K^{(e)}_{A_1,A_3}(\omega)$ in the interval of the frequencies $\omega \ll \omega_{ex}$ are presented in Fig. 6. These curves are obtained by numerical integrations of the appropriate expressions using the values of the parameters for the copper subsystem of $Nd_2CuO_4$ given in Sec. II [see (15) and the text before formula (14)]. In particular, for the dependences shown in Fig. 6 we have used the simplifying assumption that $S\pi_{12} \simeq g\mu_B$ and $I_{12}=0$. Generally speaking, in spite of the exchange nature of the $\pi_{\alpha\beta}$ tensor the values of its components may be small because, as already mentioned before, the exchange contribution to the vector of the electric-dipole moment $\mathbf{P}$ is equal to zero for the crystal with $Nd_2CuO_4$ type of magnetic symmetry if the structural distortions are absent. That means that the components of the $\pi_{\alpha\beta}$ tensor may be small by virtue of the small value of the structural distortions $\Delta_i$. The assumption $S\pi_{12} \simeq g\mu_B$ has been adopted with the only purpose to provide the possibility of comparison of the frequency dependences of the $K^{(h)}(\omega)$ and $K^{(e)}(\omega)$ coefficients in the same scale. The dependences of the absorption coefficients $K^{(h)}_{A_2,E}(\omega)$ and $K^{(e)}_{A_2,E}(\omega)$ are given in Fig. 7. The dependence of the coefficient of magnetic-dipole absorption does not follow the frequency dependence of the density of magnon states in the low-frequency region. This discrepancy between the frequency dependences of the absorption coefficient and the spin-wave density of states is caused by the structure of the $f^{(h)}_{\mu\nu}(\mathbf{k})$ amplitude as discussed above.

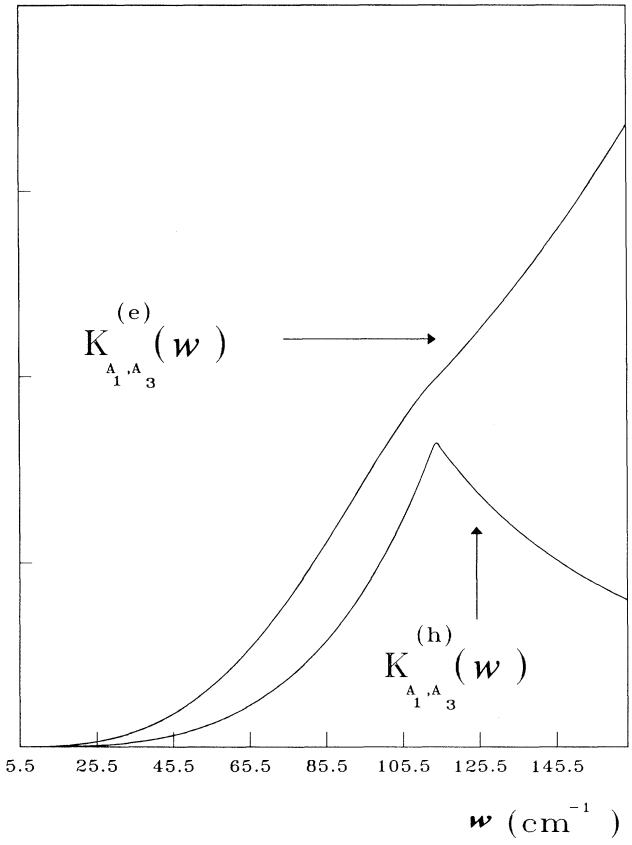


FIG. 6. Frequency dependences of the absorption coefficients $K^{(h)}_{A_1,A_3}(\omega)$ and $K^{(e)}_{A_1,A_3}(\omega)$.

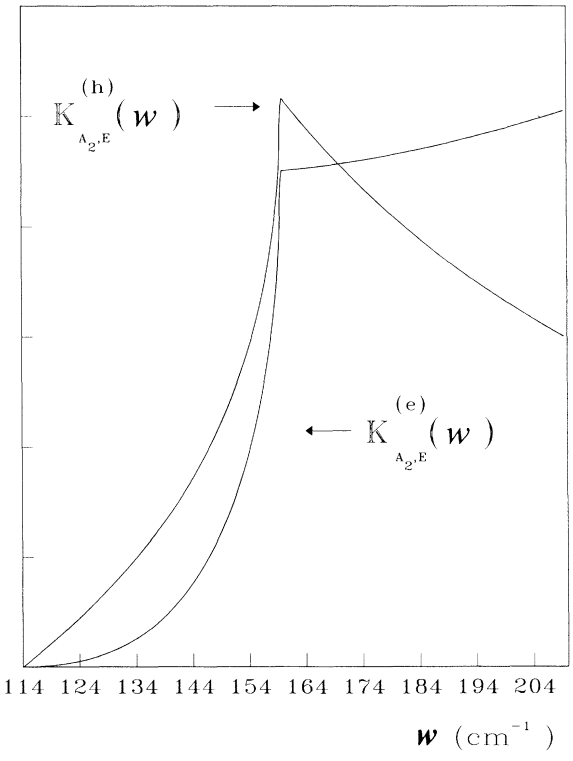


FIG. 7. Frequency dependences of the absorption coefficients $K^{(h)}_{A_2,E}(\omega)$ and $K^{(e)}_{A_2,E}(\omega)$.

To be certain, let us consider the spin-wave density of states for acoustic magnons. For the $A_3$ branch of the spin-wave spectrum, we get

$$g_{A_3}(\omega)=\frac{8\omega}{\pi\omega_{ex}^2}\arccos\left[\frac{\omega_1^2-\omega^2}{\omega_1^2-\omega_0^2}\right], \tag{32}$$

where

$$\omega_0=\omega_{A_3},\quad \omega_1=\sqrt{\omega_E^2+\omega_{A_3}^2}\ .$$

For the exchange $E$ branch of the spin-wave spectrum, we have, correspondingly,

$$g_E(\omega)=\frac{8\omega}{\pi\omega_{ex}^2}\arcsin\left[\frac{\omega^2-\omega_0^2}{\omega_1^2-\omega_0^2}\right]; \tag{33}$$

here

$$\omega_0=\sqrt{\omega_E^2+\omega_{62}^2\mathrm{sgn}(a_6-a_2)}\ ,$$
$$\omega_1=\sqrt{2\omega_E^2+\omega_{62}^2\mathrm{sgn}(a_6-a_2)}\ .$$

During the calculations of (32) and (33), the spin-wave spectrum has been expanded in the power series of the small parameters $(\mathbf{k}\cdot\hat{\mathbf{a}})\ll 1$ and $(\mathbf{k}\cdot\hat{\mathbf{b}})\ll 1$, but dispersion with respect to $k_z$ has been taken into account explicitly. The density of states $g_{A_3}(\omega)$ and $g_E(\omega)$ have square-root-type singularities at the $\omega_1$ points because these points are the saddle points for both spin-wave branches. This is a well-known result[31,32] for such kinds of the singular behavior in the spectrum of quasiparticles. The frequency dependences of the densities of states of the spin waves, $g_{A_3}(\omega)$ and $g_E(\omega)$, are given in Fig. 8. As one can see from formulas (32) and (33) and Fig. 8, the region of the nonlinear dependence of the density of states of the $A_3$ and $E$ modes on frequency ($\propto\omega_1-\omega_0$) is determined by the value of the four-site exchange interaction $D$.

The density of states for the spin-wave branches $A_1$ and $A_2$ is described by the expression

$$g_{A_1,A_2}(\omega)=\frac{8\omega}{\pi\omega_{ex}^2}\begin{cases}J & \text{at } \omega_{A_1}<\omega\le\omega_{ex},\\ 0 & \text{at } \omega\le\omega_{A_1}\ ,\end{cases} \tag{34}$$

where

$$J=\int_0^{\pi/2}\frac{d\varphi\sqrt{1-\zeta\cos\varphi}}{(1-2\zeta\cos\varphi)\sqrt{1+\zeta\cos\varphi}},\quad \zeta=\frac{I_{12}}{2I_{14}}\ .$$

It follows from formula (34) that the density of states of these modes has a step shape which has a height of the order of $4\omega_{A_1}/\omega_{ex}^2$ and increases linearly with increasing frequency. If one takes into account the dispersion of the anisotropy, the step will be smeared in a very narrow interval of the energies of the order of $\omega_0 A I_{14}^{-1}$.

Thus, in the frequency intervals corresponding to the linear dependences of the densities of states $g_{A_3}(\omega)$ and $g_E(\omega)$, the coefficients of the magnetic-dipole channel of the absorption $K_{A_1,A_3}(\omega)$ and $K_{A_2,E}(\omega)$ are inversely proportional to the square of the frequency.

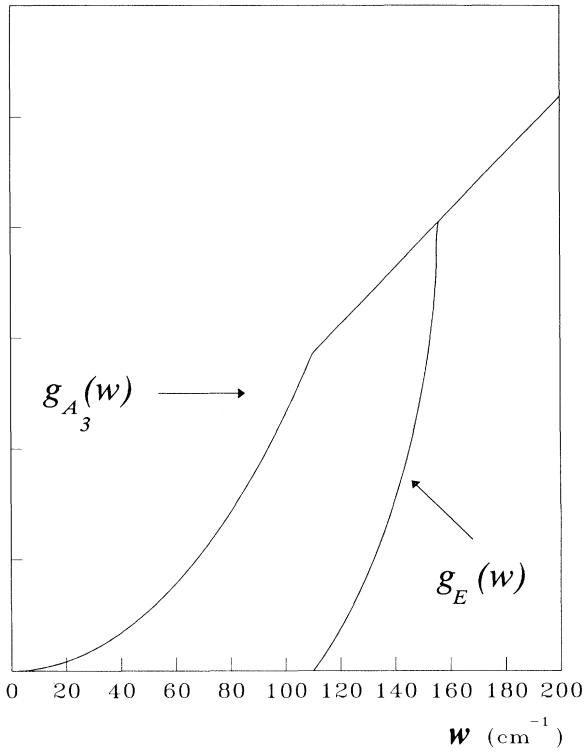


FIG. 8. Frequency dependences of the density of states for the $A_3$ acoustic and exchange branches of the spin-wave spectrum.

The main peculiarities of two-magnon absorption with the participation of the exchange mode are similar to absorption by acoustic magnons. They are as follows: The behavior of the frequency dependence of the coefficient of the absorption through the magnetic-dipole channel in the region of the frequencies near the activation energies of the exchange and acoustic magnon is nonmonotonic, while it is monotonic for the coefficient of the absorption through the electric-dipole channel in the same region. It is important to note that the peak height of magnetic-dipole absorption with the participation of the exchange mode is of the same order of magnitude as the peak height of magnetic-dipole absorption by acoustic modes. Thus two-magnon absorption by exchange modes in exchange-noncollinear magnets in the absence of an external magnetic field has a substantially larger intensity than in many-sublattice collinear magnets. It is caused by the fact that at $\omega\ll\omega_{ex}$ the heights of the step of the density of magnon states for the exchange and $A_3$ acoustic modes coincide.

Let us consider an example of the crystal in which the four-site exchange interaction is negligibly small. In this case the "plane-cross" magnetic structure is fixed by the "strong enough" relativistic interactions.[23] The expressions for the two-magnon absorption coefficients (without taking into account the $I_{12}$ exchange parameter) may be obtained from formulas (24)–(27) and (28)–(31). For absorption by the $A_1$ and $A_3$ modes, one can get

$$K^{(h)}_{A_1,A_3}(\omega)=\text{const}\frac{\pi(g\mu_B)^2}{8\omega^2}\begin{cases}\omega^4_{a_4}/\omega^2_{\text{ex}} & \text{at } \omega_0<\omega\ll\omega_{\text{ex}},\\ 0 & \text{at } \omega\le\omega_0\ ,\end{cases}$$

and

$$K^{(e)}_{A_1,A_3}(\omega)=\text{const}\times\frac{(S\pi_{12})^2}{16\omega^2}\begin{cases}[\omega^2+\omega^2_{a_4}\text{sgn}(a_4)]^2\omega^{-2}_{\text{ex}} & \text{at } \omega_0<\omega\ll\omega_{\text{ex}},\\ 0 & \text{at } \omega\le\omega_0\ ,\end{cases}\tag{35}$$

where

$$\omega_0=\omega_{A_1}+\omega_{A_3}\ .$$

In the case of absorption by the $E$ exchange and $A_2$ acoustic branches, the absorption coefficients have the form

$$K^{(h)}_{E,A_2}(\omega)=\text{const}\times\frac{\pi(g\mu_B)^2}{8\omega^2}\begin{cases}\omega^4_{a_6}/\omega^2_{\text{ex}} & \text{at } \omega_0<\omega\ll\omega_{\text{ex}},\\ 0 & \text{at } \omega\le\omega_0\ ,\end{cases}$$

$$K^{(e)}_{E,A_2}(\omega)=\text{const}\times\frac{(S\pi_{12})^2}{16\omega^2}\begin{cases}[\omega^2+\omega^2_{a_6}\text{sgn}(a_6)]^2\omega^{-2}_{\text{ex}} & \text{at } \omega_0<\omega\ll\omega_{\text{ex}},\\ 0 & \text{at } \omega\le\omega_0\ .\end{cases}\tag{36}$$

In these formulas, $\omega_0=\omega_{A_1}+\omega_{62}\text{sgn}(a_6-a_2)$.

To illustrate the influence of the interlayer exchange interaction $I_{12}$ on the frequency dependence of the magnon density of states, we will consider an example of the $A_3$ acoustic mode. In the case $D\to 0$ and for $I_{12}<0$, one can get

$$g_{A_3}(\omega)=\frac{8\omega}{\omega^2_{\text{ex}}\sqrt{1-\kappa^2}}\begin{cases}\arcsin\dfrac{(\omega^2-\omega^2_{A_3})+\omega^2_{A_3}\kappa^2}{\kappa\omega^2}-\arcsin\kappa & \text{at } \omega_{A_3}\le\omega\le\omega_{A_3}\sqrt{1+\kappa},\\ \arccos\kappa & \text{at } \omega_{A_3}\sqrt{1+\kappa}\le\omega\ll\omega_{\text{ex}}\ ,\end{cases}\tag{37}$$

where $\kappa=|I_{12}|I^{-1}_{14}$. In the same case, but for $I_{12}>0$, the density of magnon states is described by the expression

$$g_{A_3}(\omega)=\frac{8\omega}{\omega^2_{\text{ex}}\sqrt{1-\kappa^2}}\begin{cases}\dfrac{\pi}{2}+\arcsin\dfrac{\omega^2-\omega^2_{A_3}(1-\kappa^2)}{\kappa\omega^2} & \text{at } \omega_{A_3}\sqrt{1-\kappa}\le\omega\le\omega_{A_3},\\ \dfrac{\pi}{2}+\arcsin\kappa & \text{at } \omega_{A_3}\le\omega\ll\omega_{\text{ex}}\ .\end{cases}\tag{38}$$

The density of states of exchange magnons in the case $D\to 0$ is not affected by the interlayer exchange interaction

$$g_E(\omega)=\frac{8\omega}{\pi\omega^2_{\text{ex}}}\begin{cases}1 & \text{at } \omega_{62}<\omega,\\ 0 & \text{at } \omega\le\omega_{62}\ .\end{cases}$$

The peak of the coefficient of magnetic-dipole absorption remains unchanged. However, its height is determined by the ratio $(A/J)$, whereas in the case of $D\neq 0$ the peak height is of the order of $(D+A)/J$. The region of nonlinear changes in the density of magnon states $g_{A_3}(\omega)$ remains very narrow, of the order of $\omega\kappa$. In the absence of the interlayer exchange parameter $I_{12}$, the dependence of the density of states $g_{A_3}(\omega)$ also starts from the region linear with respect to $\omega$. It is necessary, however, to note that in the limit $I_{12}\to 0$ the regions of the frequencies which correspond to the first lines of the formulas (37) and (38) become unphysical and have to be omitted. A comparison of the dependences of $K^{(h)}(\omega)$ and $g(\omega)$ shows that the frequency regions in which the maximal changes of the magnetic-dipole absorption coefficient take place coincide with the regions of the linear dependence of the magnon density of states. The last circumstance justifies our neglect of the interlayer exchange interaction $I_{12}$ during the calculations.

It is necessary to note an interesting peculiarity of two-magnon absorption originating from the structure of the Hamiltonian (16). In the case when the radiation propagates along the $z$ axis, the absorption coefficient

does not depend on the orientation of the polarization of the incident electromagnetic wave with respect to the $x$ and $y$ axes. It is connected with the fulfillment of the relations for the spin-wave spectrum:

$$\omega_{A_1}(k_x,k_y,k_z)=\omega_{A_2}(k_y,k_x,k_z)\ ,$$
$$\omega_{A_3}(k_x,k_y,k_z)=\omega_{A_3}(k_y,k_x,k_z)\ . \tag{39}$$

The following equalities result from the above relations and also the explicit form of the amplitudes $f_{\mu\nu}(\mathbf{k})$:

$$K^{(h_x)}_{A_1,A_2}(\omega)=K^{(h_y)}_{A_2,A_3}(\omega),\quad K^{(e_x)}_{A_1,A_3}(\omega)=K^{(e_y)}_{A_2,A_3}(\omega)\ . \tag{40}$$

During the experimental investigations of two-magnon absorption with the scheme of the experimental setup using the fixed frequency of the generator, the absorption channels $A_1,A_3$ and $A_2,A_3$ will be indistinguishable with respect to the energy. Thus for the radiation propagating along the $z$ axis $\mathbf{k}\|oz$ and with an arbitrary polarization $h_x=h\cos\varphi$, $h_y=h\sin\varphi$, the combined absorption coefficient

$$K(\omega)=K^{(h)}_{A_1,A_3}(\omega)+K^{(e)}_{A_1,A_3}(\omega)\ ,$$

observed in experiments, does not depend on the angle $\varphi$ following from (39). The same phenomenon takes place for the case of absorption by the exchange mode $E$ because relations (39) and (40) are valid with the change $A_3\Longrightarrow E$.

## IV. CONCLUSIONS

Let us briefly summarize the main results of the present investigation.

The exchange-noncollinear antiferromagnet $Nd_2CuO_4$ may be referred to as one of the most promising magnetic crystals for the study of the manifestations of the exchange branches of the spin-wave spectrum in two-magnon absorption. The main distinguishing feature of this antiferromagnet is that there exists an electric-dipole moment associated with a pair of spins which has an exchange nature. As a result, two-magnon absorption of electromagnetic waves in $Nd_2CuO_4$ is electric-dipole active in the exchange approximation. It is necessary, however, to note that the exchange contribution to the spin-dependent electric-dipole moment is nonzero only in the case when spontaneous structural distortions are present at temperatures below $T_N$. These structural distortions have appeared as a result of the structural phase transition which takes place in $Nd_2CuO_4$ above $T_N$ (see Refs. 14 and 23). The exchange noncollinear magnetic ordering in the crystal with symmetry similar to that of $Nd_2CuO_4$ may be realized also in the case when structural distortions have not taken place. In this particular case, the exchange contribution to the electric-dipole moment will be zero because of the presence of the antitranslation operation in the magnetic structure. It is necessary, however, to note that even in this case the intensity of absorption with the participation of the exchange magnon will be of the same order of magnitude as the intensity of absorption by acoustic magnons.

In spite of its exchange nature, the contribution in question may be small because of the smallness in the structural distortions. Nevertheless, despite the small value of this exchange contribution to the electric-dipole moment or even in the case when it is absent, the intensity of the absorption process involving exchange magnons will be of the same order of magnitude as the intensity of absorption by acoustic magnons only. This last circumstance is caused by the fact that the intensity of magnetic-dipole absorption with the participation of the exchange magnon is also of the same order of magnitude as absorption by acoustic magnons. This is the main distinguishing feature of $Nd_2CuO_4$ in comparison with antiferromagnets collinear in the exchange approximation.

The analysis carried out in this paper also gives a formula for the separation of different contributions to two-magnon absorption. First, the magnetic- and electric-dipole channels can be distinguished by the proper choice of the geometry of the experiment. Second, the qualitative dependences calculated in this paper will provide a basis for the separation of the contributions of different processes into experimentally measured total absorption coefficients.

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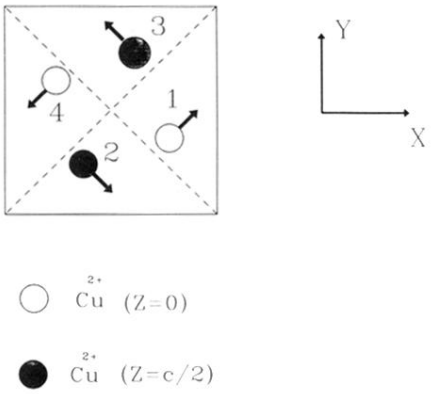


FIG. 2. Magnetic unit cell for the noncollinear magnetic phase. Solid and open circles denote the copper ions located in the $c/2$ distant crystal planes.