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Theory of Raman light scattering in the many-sublattice exchange-noncollinear magnets UO_2, $RMnO_3$, and Nd_2CuO_4 (R = rare-earth ion)

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The Raman light scattering from magnons in exchange-noncollinear many-sublattice magnets of different symmetry was studied theoretically. It is shown that in contrast to magnets collinear in the exchange approximation the intensity of scattering from the exchange magnons in exchange-noncollinear magnets is determined by the magneto-optic constants of the exchange nature, and does not contain any small factors. This intensity can be higher than the one for the acoustic magnons. The influence of the dipole-dipole interaction on the magnon states and selection rules for the Raman light scattering was investigated. Possible geometries of the light scattering from the longitudinal and transverse magnons are found.

I. INTRODUCTION

Exchange-noncollinear magnets may be referred to as many-sublattice magnetic crystals in which the spins of sublattices form a noncollinear magnetic structure in the exchange approximation. Investigations of magnetic properties of exchange-noncollinear many-sublattice magnets attract increasing interest at present.[1-18] Three examples of exchange-noncollinear magnets of different symmetry will be considered in the present study. The four-sublattice exchange-noncollinear antiferromagnet UO_2 is one of the most peculiar representatives of such magnets. A unique noncoplanar magnetic ordering when the sublattice spins are oriented along the spatial diagonals of the cubic crystal lattice takes place in this crystal. A wide variety of possible coplanar exchange-noncollinear magnetic structures may be found in the six-sublattice hexagonal perovskites with the composition $RMnO_3$ (where R is a rare-earth ion). A coplanar exchange-noncollinear magnetic structure has also been revealed in the four-sublattice antiferromagnet Nd_2CuO_4.

Exchange-noncollinear many-sublattice magnets possess, as a rule, a higher magnetic symmetry than collinear ones. The preferred direction given by the equilibrium magnetization orientation as in ferromagnets or by the equilibrium orientation of the antiferromagnetism vector as in two-sublattice antiferromagnets is absent for the magnets under consideration. Due to this circumstance it is possible to expect a more complicated and rich picture of possible spin-reorientation phase transitions in these magnets.[9-11] It was shown in Ref. 7 that the symmetry of some exchange-noncollinear antiferromagnets allows for the existence of linear magnetoelectric and piezomagnetic effects of an exchange nature.

It is necessary to note that even in magnets collinear in the exchange approximation the breaking of the collinear alignment of magnetic moments leads, as a rule, to new phenomena. This breaking of the collinearity may be caused by both the Dzyaloshinskii-Moriya interaction and the external magnetic field. For example, a contribution to the scattering tensor caused by the isotropic light scattering mechanism of the exchange nature has been revealed in antiferromagnetic EuTe in Ref. 19. The light scattering in a strong magnetic field which leads to the canting of the sublattice magnetizations has been studied in Ref. 19.

Dynamic properties of exchange-noncollinear magnets are also different from those of magnets collinear in the exchange approximation. For example, the distinguishing feature of exchange-noncollinear magnets is that the number of acoustic spin-wave modes is equal to 3 whereas for magnets collinear in the exchange approximation this number is equal to 2. The acoustic spin-wave modes may be referred to as oscillations of the sublattice spins for which the activation energy tends to zero in the exchange approximation. The type of precession of the sublattice spins during which the magnetic structure of the crystal rotates mainly as a whole, without changes of the angles between the spins forming this magnetic struc-

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ture, corresponds to the acoustic spin-wave modes. In a crystal with n magnetic sublattices the remaining $n-3$ modes of the magnetic resonance spectrum will be referred to as the exchange spin-wave modes. The activation energies of these $n-3$ modes are determined by different combinations of intersublattice exchange integrals. These activation energies remain finite in the exchange approximation. The exchange magnons are analogous to the optical phonons. During the oscillations corresponding to the exchange magnons the sublattice spins precess in antiphases and the magnetic structure of the crystal does not rotate as a whole.

It is necessary, however, to distinguish the notions of optical and exchange magnons. The term "optical magnons" has been used recently for the excitations which exist in magnetic crystals containing ions with the spin $S \geq 1$ and having a strong one-ion anisotropy.[20] These excitations correspond to transitions with nonconservation of the spin projection. The energies of these excitations may be of the order of the exchange energy. The presence or absence of the optical magnons is in no way connected with the many-sublattice structure of a magnetic crystal. They can, for example, exist even in ferromagnets which have only one magnetic ion in the unit cell. However, it is necessary to note that in earlier papers published in the 1960s and 1970s those magnons which we call the exchange ones were also called optical magnons due to their analogy with the optical phonons.

Investigation of the exchange magnons gives direct information about the values of the intersublattice exchange integrals. At the same time it provides additional data on the magnetic structure of the crystal; for example, on the angles of canting of the spins of the sublattices.

By now the exchange modes have been observed in some many-sublattice magnets by means of different experimental methods. Restricting ourselves to many-sublattice antiferromagnets, one can say that the exchange modes have been revealed in α-Fe_2O_3 by inelastic neutron scattering.[21] The exchange modes in $CsMnF_3$ manifested themselves in an indirect way by taking part in the exciton-magnon absorption.[22] Later the total spin-wave spectrum of this crystal including the exchange spin waves was studied by inelastic neutron scattering.[23] The antiferromagnetic resonance of the exchange modes has been observed in the following antiferromagnets: in the three-dimensional $CuCl_2 \cdot 2H_2O$ in Ref. 24; in the quasi-two-dimensional $(NH_3)_2(CH_2)_3MnCl_4$ in Ref. 25; in the exchange-noncollinear $CsMnBr_3$ in Ref. 5; and in a number of other antiferromagnets.

By means of the Raman light scattering method the exchange magnons have been observed only in the ferrimagnet Fe_3O_4 in Ref. 26. We shall show further that in Ref. 27, which dealt with Raman light scattering in UO_2, the scattering from the exchange magnon has actually been observed. However, the authors of Ref. 27 did not give such an interpretation to the results obtained. The known attempts to reveal the exchange magnons in the collinear antiferromagnets α-Fe_2O_3 in Ref. 28 and $NaNiF_3$ in Ref. 29 did not lead to successful results. As was theoretically shown later, the intensity of scattering from the exchange magnons caused by the ordinary mechanisms (without taking account of dissipation) is smaller than the intensity of scattering from the acoustic magnons both in three-dimensional magnets of the $R$$FeO_3$ type[30] and in quasi-two-dimensional magnets of the La_2CuO_4 type.[31] These intensities may be of the same order of magnitude only in the presence of an external magnetic field of particular value and orientation.[32] As will be clear from what follows the situation is quite different for some types of exchange-noncollinear magnets; namely, the scattering cross section for the exchange magnons may be larger than the one for the acoustic magnons. There are two reasons for this. First is that the intensity of scattering from the exchange magnons is determined by the exchange mechanism. Second is that in some cases, for example, in Nd_2CuO_4, there exists the effect of exchange enhancement of the amplitudes of oscillations corresponding to the exchange magnons.[8]

Along with the above-listed peculiarities of scattering from the exchange magnons in exchange-noncollinear magnets there will be peculiarities of scattering from the acoustic magnons. The reason is that all three acoustic magnons are magnetodipole active. This circumstance leads to such phenomena as the longitudinal-transverse splitting in the case of cubic symmetry if the dipole-dipole interaction is taken into account, or the dependence of the magnon limiting frequencies on the spin-wave propagation direction in the case of uniaxial crystals. That is why the frequency of the scattered light and the form of the scattering tensor will be dependent on the scattering geometry, similarly to the scattering from polar phonons.

II. SCATTERING TENSOR

There is a rich variety of magnons of different symmetry in many-sublattice magnets. Generally speaking, the light scattering from these magnons may not be connected with the rotation of the polarization plane. That is why in every particular case we shall be interested in the selection rules. In this connection it is convenient to consider the tensor of scattering $a_{ij}^{\nu}(\mathbf{q})$ from the magnon of the νth mode with momentum $\mathbf{q}$ instead of the scattering cross section. The Cartesian indices i and j correspond to the components of the incident and scattered light, respectively. The relative contributions of different scattering mechanisms can be easily taken into account in the framework of this approach. The scattering tensor is determined as the matrix element of the spin-dependent part of the crystal dielectric permittivity $\Delta\varepsilon_{ij}$ between the states with a difference in magnon number equal to unity:

$$a_{ij}^{\nu}(\mathbf{q})=[n_\nu(\mathbf{q})+1]^{-1/2}\langle n_\nu(\mathbf{q})+1|\Delta\varepsilon_{ij}|n_\nu(\mathbf{q})\rangle \ . \quad (1)$$

In this formula $n_\nu(\mathbf{q})$ is the number of magnons of the νth mode with momentum $\mathbf{q}$. In what follows we shall neglect the spatial dispersion of the incident and scattered light (see Ref. 20) and the wave vector $\mathbf{q}$ will only be taken into account in order to consider the effects connected with the direction of spin-wave propagation. In the same way as in Ref. 33 one can represent $\Delta\varepsilon_{ij}$ as a series in powers of the spins of the sublattices. However, in the case of the many-sublattice magnet it is very difficult to deal with the spins of separate sublattices as was done in Refs. 20 and 33. That is why we will decompose $\Delta\varepsilon_{ij}$ into a power series in linear combinations $\mathbf{L}_\rho$ of the sublattice spins entering the magnetic unit cell.[30] The components of the vectors $\mathbf{L}_\rho$ execute the irreducible representations T_ρ of the symmetry group of the paramagnetic phase of a given crystal. These linear combinations of the sublattice spins can easily be obtained with the help of the projection operator for every particular magnet. The expansion for $\Delta\varepsilon_{ij}$ may be written as

$$\Delta\varepsilon_{ij}=\sum \lambda_{ijl}^{(\rho)} L_{\rho l}+\sum \sigma_{ijlm}^{(\rho\delta)} L_{\rho l} L_{\delta m}+\sum P_{ij}^{(\rho\delta)}(\mathbf{L}_\rho \cdot \mathbf{L}_\delta) . \quad (2)$$

The first term in expression (2) describes the Faraday effect if one substitutes the equilibrium values $\overline{\mathbf{L}}_\rho$ for the operators $\mathbf{L}_\rho$ in the absence of absorption. The two other terms describe the Cotton-Mouton effect under the same conditions. The form of the tensors $\hat{\lambda}$, $\hat{\sigma}$, and $\hat{P}$ can easily be determined because the values $\mathbf{L}_\rho$ are transformed in accordance with the irreducible representations of the symmetry group of the paramagnetic phase of a given magnet.[30] The values $\hat{\sigma}$ and $\hat{P}$ can be expressed through the polarizability of the pair of ions if required. The last term in expression (2) has exchange nature. We shall not be interested further in the microscopic nature of the values $\hat{\lambda}$, $\hat{\sigma}$, and $\hat{P}$, which have been considered in detail in Refs. 33–35. The main attention will be paid to the elucidation of the role and value of the contribution of the exchange mechanism, which is determined by the constant P, in the scattering of light from exchange and acoustic magnons in exchange-noncollinear magnets.

The form of the scattering tensor $a_{ij}^{\nu}(\mathbf{q})$ depends on what components of the vectors $\mathbf{L}_\rho$ take part in the oscillations corresponding to the given mode ν. The classification of the uniform oscillations of the spin system and the calculations of the energies of these oscillations will be carried out for every particular magnet separately. In order to compare the relative contributions of the values λ, σ, and P to the components of the scattering tensor $a_{ij}^{\nu}(\mathbf{q})$ we have to obtain the relative amplitudes of the oscillations of the components of the $\mathbf{L}_\rho$ vectors for every magnon mode. These amplitudes can be expressed through the coefficients of the unitary u-v transformation in the scheme of the second quantization of the Hamiltonian of the many-sublattice magnet[30] in the simplest way. The method of the second quantization of the Hamiltonian of the many-sublattice magnet developed in Ref. 30 allows one to take into account the symmetry of the magnet in the most comprehensive way and will be used in all the cases under investigation.

III. MAGNETIC STRUCTURE OF THE UO_2 TYPE

The four-sublattice exchange-noncollinear noncoplanar magnetic structure of UO_2 is shown in Fig. 1. The symmetry of the paramagnetic phase of this crystal is described by the $Fm3m$ group. There is one chemical formula unit in the primitive cell. The magnetic unit cell is four times larger than the crystallographic one. The magnetic symmetry group is $Pn3m'$. The consideration will be carried out in the framework of conventional spin-wave theory for $S \gg 1$ at $\mathbf{k}=\mathbf{0}$ and for the temperature region $T \ll T_N$. The applicability of this approach will be discussed later.

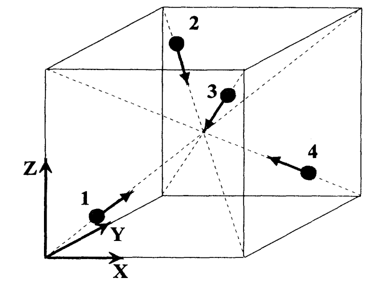

FIG. 1. Magnetic structure of UO_2.

The possibility of the existence of such a magnetic structure has been pointed out in Ref. 36. Following this paper, let us introduce the linear combinations of the sublattice spins

$$\begin{aligned}&\mathbf{F}=\mathbf{S}_1+\mathbf{S}_2+\mathbf{S}_3+\mathbf{S}_4, \quad \mathbf{L}_1=\mathbf{S}_1+\mathbf{S}_2-\mathbf{S}_3-\mathbf{S}_4 ,\\ &\mathbf{L}_2=\mathbf{S}_1-\mathbf{S}_2+\mathbf{S}_3-\mathbf{S}_4, \quad \mathbf{L}_3=\mathbf{S}_1-\mathbf{S}_2-\mathbf{S}_3+\mathbf{S}_4 .\end{aligned} \quad (3)$$

From the point of view of the high-symmetry (paramagnetic) phase the magnetic ordering in UO_2 is caused by the phase transition corresponding to the three-ray channel of the irreducible star $\mathbf{k}_{10}$ (here and further the notation of Ref. 37 is used) of the $Fm3m$ group with the following rays:

$$\mathbf{k}_1=\tfrac{1}{2}(\mathbf{b}_2+\mathbf{b}_3), \quad \mathbf{k}_2=\tfrac{1}{2}(\mathbf{b}_1+\mathbf{b}_3), \quad \mathbf{k}_3=\tfrac{1}{2}(\mathbf{b}_1+\mathbf{b}_2) .$$

Here $\mathbf{b}_1$, $\mathbf{b}_2$, and $\mathbf{b}_3$ are the vectors of the reciprocal lattice. The vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ are parallel to the X, Y, and Z axes, respectively (see Fig. 1). The components of the vectors $\mathbf{L}_1$, $\mathbf{L}_2$, and $\mathbf{L}_3$ correspond to the one-dimensional irreducible representation T_3 and the two-dimensional irreducible representation T_9 of the group of the wave vector of the ray of the star $\mathbf{k}_{10}$ (see Table I). The F_x, F_y, and F_z components of the vector $\mathbf{F}$ belong to the same three-dimensional even representation T_9 of the star $\mathbf{k}_{11}(\mathbf{k}=\mathbf{0})$. The following components of the vectors $\mathbf{L}_\rho$ are nonzero in the ground state:

$$\overline{L}_{1x}=\overline{L}_{2y}=\overline{L}_{3z}=4S/\sqrt{3} . \quad (4)$$

Using all the aforesaid the spin-dependent part of the dielectric permittivity of the crystal can be represented in the form

TABLE I. Distribution of the components of the vectors $\mathbf{L}$ with respect to the irreducible representations of the group of the wave vector of the ray of the star $\mathbf{k}_{10}$.

Irreducible representation	$\mathbf{k}_1$	$\mathbf{k}_2$	$\mathbf{k}_3$
T_3	L_{1x}	L_{2y}	L_{3z}
T_9	$\begin{cases} L_{1y} \\ L_{1z} \end{cases}$	$\begin{cases} L_{2z} \\ L_{2x} \end{cases}$	$\begin{cases} L_{3x} \\ L_{3y} \end{cases}$

$$\begin{aligned}
\Delta\varepsilon_{xx} &= P_1\mathbf{L}_1^2+P_2\mathbf{F}^2+\sigma_1 L_{1x}^2 \\
&\quad +\sigma_2(L_{1x}^2+L_{2y}^2+L_{3z}^2)+\sigma_3 F_x^2\ , \\
\Delta\varepsilon_{yy} &= P_1\mathbf{L}_2^2+P_2\mathbf{F}^2+\sigma_1 L_{2y}^2 \\
&\quad \sigma_2(L_{1x}^2+L_{2y}^2+L_{3z}^2)+\sigma_3 F_y^2\ , \\
\Delta\varepsilon_{zz} &= P_1\mathbf{L}_3^2+P_2\mathbf{F}^2+\sigma_1 L_{3z}^2 \\
&\quad +\sigma_2(L_{1x}^2+L_{2y}^2+L_{3z}^2)+\sigma_3 F_z^2\ , \\
\Delta\varepsilon_{xy} &= \lambda F_z+\sigma_4(L_{1x}L_{1y}+L_{2x}L_{2y}) \\
&\quad +\sigma_5 L_{3x}L_{3y}+\sigma_6 F_x F_y\ , \\
\Delta\varepsilon_{zx} &= \lambda F_y+\sigma_4(L_{1x}L_{1z}+L_{3x}L_{3z}) \\
&\quad +\sigma_5 L_{2x}L_{2z}+\sigma_6 F_x F_z\ , \\
\Delta\varepsilon_{yz} &= \lambda F_x+\sigma_4(L_{2y}L_{2z}+L_{3y}L_{3z}) \\
&\quad +\sigma_5 L_{1y}L_{1z}+\sigma_6 F_y F_z\ .
\end{aligned} \tag{5}$$

In order to investigate the scattering tensor obtained with the help of (5) one has to eliminate the terms linear with respect to the operators of creation and annihilation of the magnons. For this we consider the following Hamiltonian corresponding to the given magnetic structure:[5]

$$\mathcal{H}=\tilde{B}\mathbf{F}^2-\tilde{a}(L_{1x}^2+L_{2y}^2+L_{3z}^2)+\tilde{D}(\mathbf{L}_1^4+\mathbf{L}_2^4+\mathbf{L}_3^4)\ . \tag{6}$$

In this expression $\tilde{B}$ is the exchange constant, $\tilde{a}$ is the constant of magnetic anisotropy, and $\tilde{D}$ describes the biquadratic exchange. The constants describing the contributions of different interactions to the Hamiltonian have the dimension of energy throughout this paper. We shall consider the case $\hbar=1$, that is, the energy and frequency have the same dimension. Writing expression (6) we used the relations

$$\begin{gathered}
\mathbf{F}^2+\mathbf{L}_1^2+\mathbf{L}_2^2+\mathbf{L}_3^2=(4S)^2\ , \\
\mathbf{L}_1\cdot\mathbf{L}_2+\mathbf{F}\cdot\mathbf{L}_3=\mathbf{L}_1\cdot\mathbf{L}_3+\mathbf{F}\cdot\mathbf{L}_2=\mathbf{L}_2\cdot\mathbf{L}_3+\mathbf{F}\cdot\mathbf{L}_1=0\ .
\end{gathered} \tag{7}$$

The only fourth-order invariants ($\mathbf{L}_p^4$) which determine the static and dynamic properties of the magnet under consideration are retained in expression (6).

The conditions of stability of the magnetically ordered phase (4) have the form[2] $\tilde{a}>0$, $\tilde{D}>0$. The spin-wave spectrum and classification of the oscillations in accordance with the symmetry types have been determined in Ref. 38 with the help of the linearized equations of motion for the operators $\mathbf{L}$. Below we shall reproduce these results in the framework of the second-quantization method[30] using the Holstein-Primakoff representation. To do this one has to transform the operators $\mathbf{S}_\alpha$ into the operators $\mathbf{S}'_\alpha$ each written in its own local coordinate system with the $0z'$ axis oriented along the equilibrium direction of the spins. The matrices of the transformation $\hat{p}$ for different ions which can be permuted by the symmetry operations are connected with each other. That is why one can find the relations between the operators $\mathbf{L}$ and new operators $\mathbf{L}'$ which are the linear combinations of the operators $\mathbf{S}'_\alpha$ of the form given in (3)

$$\begin{aligned}
&\begin{bmatrix} F_x \\ L_{3y} \\ L_{2z} \end{bmatrix}_{A_1} = \hat{p}\begin{bmatrix} L'_{1x} \\ L'_{1y} \\ L'_{1z} \end{bmatrix}, \quad
\begin{bmatrix} L_{3x} \\ F_y \\ L_{1z} \end{bmatrix}_{A_2} = \hat{p}\begin{bmatrix} L'_{2x} \\ L'_{2y} \\ L'_{2z} \end{bmatrix}, \\
&\begin{bmatrix} L_{2x} \\ L_{1y} \\ F_z \end{bmatrix}_{A_3} = \hat{p}\begin{bmatrix} L'_{3x} \\ L'_{3y} \\ L'_{3z} \end{bmatrix}, \quad
\begin{bmatrix} L_{1x} \\ L_{2y} \\ L_{3z} \end{bmatrix}_{E} = \hat{p}\begin{bmatrix} F'_x \\ F'_y \\ F'_z \end{bmatrix}.
\end{aligned} \tag{8}$$

As will be shown in what follows, the subscripts of every column in the left-hand sides of equalities (8) are the numbers of magnon modes. The components of the vectors $\mathbf{L}$ entering the νth column take part in the oscillations corresponding to this mode ν. The matrix $\hat{p}$ in (8) is the matrix of the transformation into the local coordinate system of the first ion (see Fig. 1). This matrix has the form

$$\hat{p}=\begin{bmatrix} 6^{-1/2} & -2^{-1/2} & 3^{-1/2} \\ 6^{-1/2} & 2^{-1/2} & 3^{-1/2} \\ -(\frac{2}{3})^{1/2} & 0 & 3^{-1/2} \end{bmatrix}. \tag{9}$$

The $0Z'$ axis of the local coordinate system is directed along [111] and the $0Y'$ axis is oriented along the direction $[\bar{1}10]$ in the XY plane.

The operators $\mathbf{L}'$ are directly expressed in terms of linear combinations of the Holstein-Primakoff spin-deflection operators $b_\alpha^\dagger$ and b_α of separate sublattices ($\alpha=1,2,3,4$)

$$\begin{aligned}
&L'_x=\sqrt{4S}\,Q_L,\quad L'_y=i\sqrt{4S}\,P_L,\quad L=\{F,L_1,L_2,L_3\}\ ; \\
&F'_z=4S+\tfrac{1}{2}\sum_L\{P_L^2-Q_L^2\}\ ; \\
&L'_{1z}=P_F P_{L_1}+P_{L_2}P_{L_3}-Q_F Q_{L_1}-Q_{L_2}Q_{L_3}\ ; \\
&L'_{2z}=L'_{1z}\{L_1\Longleftrightarrow L_2\},\quad L'_{3z}=L'_{1z}\{L_1\Longleftrightarrow L_3\}\ .
\end{aligned} \tag{10}$$

In these expressions

$$\begin{aligned}
&Q_L=\frac{1}{\sqrt{2}}\{\beta_L^\dagger+\beta_L\},\quad P_L=\frac{1}{\sqrt{2}}\{\beta_L^\dagger-\beta_L\}\ ; \\
&\beta_F=\tfrac{1}{2}[b_1+b_2+b_3+b_4],\quad \beta_{L_1}=\tfrac{1}{2}[b_1+b_2-b_3-b_4]\ ; \\
&\beta_{L_2}=\tfrac{1}{2}[b_1-b_2+b_3-b_4],\quad \beta_{L_3}=\tfrac{1}{2}[b_1-b_2-b_3+b_4]\ .
\end{aligned} \tag{11}$$

The operators β_L are determined in such a way that the ordinary commutation relations

$$[\beta_L,\beta_{L'}^\dagger]=\delta_{LL'} \tag{12}$$

are satisfied. The operators Q_L and P_L in (10) are analogous to the generalized coordinates and momenta for some oscillators. Substituting (8)–(10) into expression (6), one can get the part of the Hamiltonian quadratic with respect to the Q_L and P_L operators,

$$\mathcal{H}^{(2)}=\frac{1}{2}\left\{\frac{1}{\sqrt{3}}[B+3a](Q_{L_1}^2+Q_{L_2}^2)\right.$$

$$+\sqrt{3}[B+a](P_{L_1}^2+P_{L_2}^2)$$

$$-iB(P_{L_1}Q_{L_1}-P_{L_2}Q_{L_2})+\frac{1}{\sqrt{3}}[4B+3a]Q_{L_3}^2$$

$$\left.-a\sqrt{3}P_{L_3}^2+D(Q_F^2-P_F^2)\right\} . \tag{13}$$

In this expression

$$B=\frac{4S}{\sqrt{3}}[\tilde{B}-\tfrac{2}{3}(4S)^2\tilde{D}],\quad a=\frac{8S}{\sqrt{3}}\tilde{a},\quad D=\tfrac{8}{3}(4S)^3\tilde{D} .$$

Using the unitary transformation

$$\begin{aligned} Q_L&=\frac{1}{\sqrt{2}}\sum_\nu(t_{L\nu}^*\xi_\nu^\dagger+t_{L\nu}\xi_\nu) ,\\ P_L&=\frac{1}{\sqrt{2}}\sum_\nu(d_{L\nu}^*\xi_\nu^\dagger-d_{L\nu}\xi_\nu) , \end{aligned} \tag{14}$$

one can represent Hamiltonian (13) in the standard form

$$\mathcal{H}^{(2)}=\sum_\nu \varepsilon_\nu\xi_\nu^\dagger\xi_\nu . \tag{15}$$

Note that the summation over the wave vectors of the magnons is absent in expression (15) because we consider the case when the wave vector of spin waves is negligibly small. That is why all the calculations have been carried out for $\mathbf{k}=\mathbf{0}$. In correspondence with the results of Ref. 38 the spin-wave spectrum of the crystal under consideration contains three acoustic magnons which are threefold degenerate with respect to the energy. These magnons have the energies

$$\varepsilon_{A_1,A_2,A_3}=\varepsilon_A=[a(4B+3a)]^{1/2} .$$

There is also one exchange magnon for which the activation energy is determined by the biquadratic exchange

$$\varepsilon_E=D .$$

Thus the meaning of the classification (8) is the following: those components of the vectors $\mathbf{L}$ take part in the oscillations of the same magnon mode which contain terms linear in the coordinate or momentum of the same oscillator.

The coefficients of the unitary transformation $t_{L\nu}=u_{L\nu}+v_{L\nu}$ and $d_{L\nu}=u_{L\nu}-v_{L\nu}$ have the form

$$\begin{aligned} t_{L_1A_1}&=3^{1/4}\frac{\varepsilon_A-iB}{[\varepsilon_A(B+3a)]^{1/2}} ,\\ d_{L_1A_1}&=3^{-1/4}\left[\frac{B+3a}{\varepsilon_A}\right]^{1/2} ,\\ t_{L_2A_2}&=3^{1/4}\frac{\varepsilon_A+iB}{[\varepsilon_A(B+3a)]^{1/2}} ,\\ d_{L_2A_2}&=3^{-1/4}\left[\frac{B+3a}{\varepsilon_A}\right]^{1/2} ,\\ t_{L_3A_3}&=-\left[\frac{3a}{4B+3a}\right]^{1/4} ,\\ d_{L_3A_3}&=-\left[\frac{4B+3a}{3a}\right]^{1/4} ,\\ t_{F\nu_E}&=d_{F\nu_E}=1 . \end{aligned} \tag{16}$$

It follows from (16) that all the coefficients $d_{L\nu}$ are exchange enhanced, that is, they are of the order of $(B/a)^{1/4}$. The substitution of (14) and (16) into (8) allows one to analyze the relative amplitudes for all magnon modes. Thus for the oscillations corresponding to the acoustic magnons the components of the vector $\mathbf{F}$ have the smallest relative amplitudes and those components of the vectors $\mathbf{L}_1$, $\mathbf{L}_2$, and $\mathbf{L}_3$ for which the equilibrium values are equal to zero have the largest relative amplitudes. The last circumstance reflects the Goldstone character of the acoustic magnons in the framework of the Hamiltonian considered. In the exchange approximation the rotations of the magnetic structure as a whole around the X, Y, and Z axes correspond to the oscillations of the modes A_1, A_2, and A_3 with $\mathbf{k}=\mathbf{0}$, respectively. The oscillations corresponding to the exchange mode cannot be reduced to a uniform rotation of the magnetic structure as a whole.

Using the obtained expressions and relations (1) and (5) one can determine the scattering tensors for all magnon branches. For the acoustic magnons these tensors are the following:

$$\begin{aligned} a_{ij}^{A_1}&=\begin{bmatrix}0&0&0\\0&0&Ge^{-i\delta}\\0&Re^{-i\delta}&0\end{bmatrix} ,\\ a_{ij}^{A_2}&=\begin{bmatrix}0&0&Re^{i\delta}\\0&0&0\\Ge^{i\delta}&0&0\end{bmatrix} ,\quad a_{ij}^{A_3}=\begin{bmatrix}0&G&0\\R&0&0\\0&0&0\end{bmatrix} , \end{aligned} \tag{17}$$

where

$$R=\left[\frac{4S}{\sqrt{3}}\right]^{1/2}\left[\frac{a}{4B+3a}\right]^{1/4}\left[i\lambda+\frac{4S}{\sqrt{3}}\sigma_4\right] ,$$

$$G=R^*,\quad e^{i\delta}=\frac{(4B+3a)^{1/2}+i3\sqrt{a}}{2(B+3a)^{1/2}} .$$

Let us note two circumstances following from expressions (17). First, both the antisymmetric term (the Faraday term) and the symmetric term (proportional to σ_4) are exchange weakened by the factor $(a/B)^{1/4}$. The symmetric term of relativistic origin is exchange enhanced in some cases in collinear antiferromagnets.[20,30] Second, the calculations show that the Stokes–anti-Stokes asymmetry of the scattering intensity[20] which is usually observed in collinear magnets will be absent in the case under consideration. Thus the scattering from the acoustic magnons in collinear and noncollinear antiferromagnets, being different in detail, have one common feature that the contribution of the Faraday term is always exchange weakened.

An absolutely different situation takes place during the scattering from the exchange magnons. The scattering tensor for this case has the form

$$a_{ij}^{\nu_E}=\begin{bmatrix} Pe^{i2\pi/3} & 0 & 0 \\ 0 & Pe^{-i2\pi/3} & 0 \\ 0 & 0 & P \end{bmatrix} , \tag{18}$$

where $P=(8S/3)(4S)^{1/2}P_1$. As follows from (18) the given tensor does not contain any small factors and is determined by the quadratic magneto-optic constant of the exchange nature. The quantity P_1 can be expressed in terms of the spin-dependent exchange part of the polarizability of the pair of ions. The exchange part of the polarizability of the pair of ions may be written as $\alpha_{ij}^{(\mathrm{ex})}=\mathrm{Tr}\{\alpha_{ijll}\}$, where the trace is taken with respect to the spin Cartesian indices. The expression for P_1 is

$$P_1=\alpha_{\perp}^{(\mathrm{ex})}-\alpha_{\parallel}^{(\mathrm{ex})} .$$

In this expression $\alpha_{\parallel}^{(\mathrm{ex})}$ and $\alpha_{\perp}^{(\mathrm{ex})}$ are the components of the polarizability of a pair of neighboring ions longitudinal and transverse with respect to the line connecting these ions. As could be expected the form of tensors (17) and (18) coincides with the ones found in Ref. 39 for the magnetic point group $m3m'$. In the spectroscopic notation the tensors (17) describe the scattering from the T_g modes and tensor (18) describes the scattering from the E_g mode. It has to be noted here that the two-dimensional corepresentation of the Shubnikov group $m3m'$ corresponds to the representation E_g of the unitary subgroup. This two-dimensional corepresentation of the Shubnikov group $m3m'$ contains two combined complex-conjugate one-dimensional representations, one of which corresponds to the nonphysical value of the energy $-\varepsilon_E$.

Let us consider the peculiarities of the manifestation of the dipole-dipole interaction in the light scattering from the acoustic magnons. The acoustic magnon with wave vector $\mathbf{q}=\mathbf{k}_i-\mathbf{k}_s$ ($\mathbf{k}_i$ and $\mathbf{k}_s$ are the wave vectors of the incident and scattered light) is created or annihilated during the scattering process. If the so-called "forward-scattering" geometry is not considered we have that $|\mathbf{q}|\simeq|\mathbf{k}_i|,|\mathbf{k}_s|$ for all the other geometries of the scattering. The inequality $\mathcal{L}^{-1}\ll q\ll a_0^{-1}$, where $\mathcal{L}$ is the sample size and a_0 is the lattice constant, is as a rule satisfied for such wave vectors. In this case on the one hand one can neglect the lag effects (the polariton effects) because $\hbar\varepsilon_\nu/c\ll q$ and on the other hand the dispersion in the magnon spectrum can still be neglected. As was already mentioned in Sec. II the magnon wave vector will be considered in order to take into account the effects depending on the spin-wave propagation direction only. In order to determine the dependence of the energy of the magnon on its propagation direction one has to add a term describing the nonanalytic part of the dipole-dipole interaction to the Hamiltonian (15). This term may be written as

$$\mathcal{H}_{d\text{-}d}=\frac{2\pi}{v_0}(g\mu_B)^2\frac{(\mathbf{q}\cdot\mathbf{F})^2}{|\mathbf{q}|^2} . \tag{19}$$

In this expression v_0 is the volume of the unit cell, g is the g factor, and μ_B is the Bohr magneton. Note that the zero Fourier component of the vector $\mathbf{F}$ enters expression (19). The analytic part of the dipole-dipole interaction will not be considered because it does not depend on the direction of the spin-wave propagation.

Separating out the part of (19) which is quadratic with respect to the operators of creation and annihilation of magnons and carrying out the diagonalization procedure for the total Hamiltonian $\mathcal{H}^{(2)}+\mathcal{H}_{d\text{-}d}^{(2)}$ once again, one can get the magnon energies. We obtained the following expressions for the energies of the "transverse" acoustic magnons:

$$\Omega_{\perp 1}(\mathbf{q})=\Omega_{\perp 2}(\mathbf{q})=\varepsilon_A , \tag{20}$$

and for the "longitudinal" acoustic magnon

$$\Omega_{\parallel 1}(\mathbf{q})=\left[a\left[4B+3a+\frac{32S\pi}{v_0\sqrt{3}}(g\mu_B)^2\right]\right]^{1/2} . \tag{21}$$

As one can see from (21) the value of the longitudinal-transverse splitting is small because the dipole-dipole contribution enters the magnon energy as an addition to the large exchange constant. The terms "longitudinal" and "transverse" acoustic magnons characterize the direction of the oscillation of the magnetization with respect to the magnon wave vector $\mathbf{q}$. In particular, the components of the operator of the total spin of the magnetic cell assume the following form after the transition to the operators $\xi_\rho^\dagger$ and ξ_ρ of creation and annihilation of the magnons:

$$\hat{F}_i=\left[\frac{4S}{\sqrt{3}}\right]^{1/2}\left[\frac{a}{4B+3a}\right]^{1/4} \times\left\{\mathbf{e}_{\perp 1,i}(\xi_{\perp 1}^\dagger+\xi_{\perp 1})+\mathbf{e}_{\perp 2,i}(\xi_{\perp 2}^\dagger+\xi_{\perp 2}) +\left[\frac{\Omega_\parallel}{\Omega_\perp}\right]^{1/2} f_i(\xi_\parallel^\dagger+\xi_\parallel)\right\} . \tag{22}$$

Here the vectors of the magnon polarization $\mathbf{e}_{\perp 1},\mathbf{e}_{\perp 2}$, and $\mathbf{f}=\mathbf{q}\cdot|\mathbf{q}|^{-1}$ form the three mutually orthogonal unit vectors, with the vector $\mathbf{e}_{\perp 1}$ lying in the X,Y plane.

Now the form of the tensor of scattering from the acoustic magnons is determined by the relation

$$a_{ij}^{\rho}=\sum_\nu l_{\rho\nu}a_{ij}^{\nu} , \tag{23}$$

where the coefficients $l_{\rho\nu}$ are determined by the expression

$$\begin{Bmatrix} l_{\rho A_1} \\ l_{\rho A_2} \\ l_{\rho A_3} \end{Bmatrix} = \begin{Bmatrix} e^{i\delta} e_{\rho,x} \\ e^{-i\delta} e_{\rho,x} \\ e_{\rho,z} \end{Bmatrix} , \quad (24)$$

in which the notation $\mathbf{e}_{\parallel} = \mathbf{f}$ is used. The quantity δ is determined after formulas (17).

The light scattering from all types of magnons will be observed in the 90° geometry of scattering. However, in the case of the "backward-scattering" geometry the only possible scattering process is with the participation of the longitudinal acoustic magnon as follows from relations (17) and (23).

The magnetic-ordering phase transition in UO_2 takes place as a first-order phase transition. Without going into discussion of the mechanism of this phenomenon, it is necessary to remark that there is a subgroup relation between the high- and low-symmetry phases. This circumstance allows one to use the above-given terminology characteristic for a second-order phase transition for the derivation of the Hamiltonian. The second circumstance consists in that the ground state of the U^{4+} ion is described by the effective spin $S = 1$. The magnetic spectrum contains "spin" and "quadrupole" excitations.[40] However, the analysis of the spin-wave representation carried out by us shows that the symmetry of the spin excitations is retained the same as in the above-considered model. Thus the results obtained here may be applied to the analysis of the light scattering from the spin excitations in UO_2 at $T \ll T_N$ in this qualitative sense.

The consideration of the data of Ref. 27 is of great interest from this point of view. The Raman light scattering in the backward-scattering geometry has been experimentally investigated in the antiferromagnetic phase of UO_2. Scattering from magnetic excitations with symmetry E_g and the value of the energy equal to 18.5 cm^{-1} and also with symmetry T_g and the energies 78 and 98 cm^{-1} was observed. The magnetic character of these excitations has been proved by the fact that their intensity tended to zero as the temperature increased towards the Néel temperature. Another confirmation is the agreement of the observed values of the energies with the data obtained by inelastic neutron scattering.[41] Such experimental facts as the number of the lines and their symmetry were in contradiction with the theory used by the authors of Ref. 27 for the interpretation of the results. The theory used was based on the two-sublattice collinear antiferromagnetic model of UO_2.

Note that the symmetry of the magnetic excitations revealed in Ref. 27 and the circumstance that the excitation with energy 18.5 cm^{-1} and symmetry E_g is not manifested in the infrared absorption are already enough to make a conclusion that this excitation is the exchange magnon. They are also enough to conclude that the magnetic structure of UO_2 is an exchange-noncollinear noncoplanar magnetic structure. The calculations of the magnon energies made later in Ref. 40 in the framework of the model of the exchange-noncollinear noncoplanar magnetic structure taking into account the effective spin value $S = 1$ have shown that the exchange spin magnon does have smaller energy than the acoustic spin magnons (having energies equal to 80 cm^{-1}) and the acoustic quadrupole magnons (with energy 100 cm^{-1}). The data of inelastic neutron scattering[41,42] were used to determine the constants of the Hamiltonian of UO_2 in Ref. 40. The symmetry classification of the magnon branches was not carried out in Ref. 40. By this means the results of Ref. 27 could serve as a proof of the fact that the magnetic structure of UO_2 is noncollinear and noncoplanar already in 1975.

As one can clearly see from the spectrum given in Ref. 27 the intensity of the line of the scattering from the exchange magnon is actually higher than the intensity of the lines of the scattering from the acoustic magnons. This result is the remarkable circumstance for our theory considered here.

IV. MAGNETIC STRUCTURE OF THE *R*MnO_3 TYPE

There is a big group of exchange-noncollinear coplanar magnets with the so-called triangular magnetic ordering. Compounds with the composition $RMnO_3$ where R is a rare-earth ion are representative of this group. The symmetry of the paramagnetic phase of these compounds is described by the spatial group C_{6v}^3-$P6_3cm$. The primitive cell contains six formula units of $RMnO_3$. The manganese ions occupy the 6c positions. There are three of them in every plane normal to the sixfold symmetry axis and separated from each other by a distance equal to half of the lattice period along the hexagonal axis. We consider the Raman light scattering from magnons for two possible types of magnetic ordering which are shown in Figs. 2 and 3. The appearance of these magnetic structures is not accompanied by the multiplication of the primitive crystal cell. Consequently, there are three acoustic and three exchange magnons in these cases. The symmetry, spin waves, and peculiarities of magnetoelastic properties for both these types of magnetic ordering in $RMnO_3$ have been considered in Ref. 3. Following this paper, we also introduce the linear combinations of the

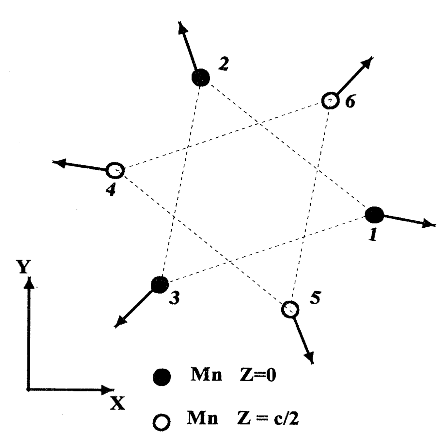

FIG. 2. Magnetically ordered phase T_2 of $RMnO_3$.

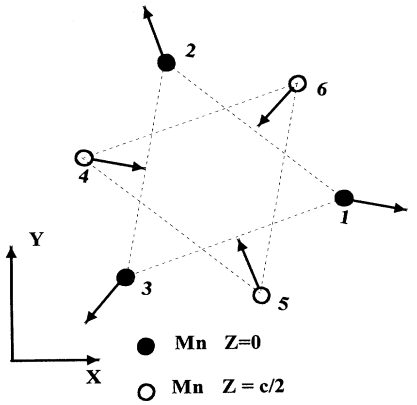

FIG. 3. Magnetically ordered phase T_3 of $R\mathrm{MnO}_3$.

vectors of spins of the ions. These linear combinations execute the permutational representation and in our case they are as follows:

$$\begin{aligned}
&\varphi_1=\mathbf{S}_1+\mathbf{S}_2+\mathbf{S}_3+\mathbf{S}_4+\mathbf{S}_5+\mathbf{S}_6\ ,\\
&\varphi_4=\mathbf{S}_1+\mathbf{S}_2+\mathbf{S}_3-\mathbf{S}_4-\mathbf{S}_5-\mathbf{S}_6\ ,\\
&\varphi_{5,\mathrm{I}}=\mathbf{S}_1-\omega^*\mathbf{S}_2-\omega\mathbf{S}_3-\mathbf{S}_4+\omega^*\mathbf{S}_5+\omega\mathbf{S}_6\ ,\\
&\varphi_{5,\mathrm{II}}=-\varphi_{5,I}^*\ ,\\
&\varphi_{6,\mathrm{I}}=\mathbf{S}_1-\omega^*\mathbf{S}_2-\omega\mathbf{S}_3+\mathbf{S}_4-\omega^*\mathbf{S}_5-\omega\mathbf{S}_6\ ,\\
&\varphi_{6,\mathrm{II}}=\varphi_{6,\mathrm{I}}^*\ .
\end{aligned}\tag{25}$$

In these expressions the subscript of the linear combination denotes the number of the irreducible representation of the group in the framework of the numbering scheme adopted in Ref. 37; the roman numerals number the linear combinations for two-dimensional representations; $\omega=\exp\{i\pi/3\}$. The form of the linear combinations (25) is slightly different from the ones used in Ref. 3 and has been chosen for convenience of performance of the second quantization. The following unimodularity conditions are satisfied for vectors (25):

$$\begin{aligned}
&\varphi_1^2+\varphi_4^2-2\varphi_{5,\mathrm{I}}\cdot\varphi_{5,\mathrm{II}}+2\varphi_{6,\mathrm{I}}\cdot\varphi_{6,\mathrm{II}}=36S^2\ ,\\
&\varphi_1\cdot\varphi_4+\varphi_{5,\mathrm{I}}\cdot\varphi_{6,\mathrm{II}}-\varphi_{6,\mathrm{I}}\cdot\varphi_{5,\mathrm{II}}=0\ ,\\
&\varphi_1\cdot\varphi_{5,\mathrm{I}}+\varphi_4\cdot\varphi_{6,\mathrm{I}}-\varphi_{5,\mathrm{II}}\cdot\varphi_{6,\mathrm{II}}=0\ ,\\
&\varphi_1\cdot\varphi_{5,\mathrm{II}}-\varphi_4\cdot\varphi_{6,\mathrm{II}}-\varphi_{5,\mathrm{I}}\cdot\varphi_{6,\mathrm{I}}=0\ ,\\
&\varphi_1\cdot\varphi_{6,\mathrm{I}}+\varphi_4\cdot\varphi_{5,\mathrm{I}}+\varphi_{5,\mathrm{II}}^2+\varphi_{6,\mathrm{II}}^2=0\ ,\\
&\varphi_1\cdot\varphi_{6,\mathrm{II}}-\varphi_4\cdot\varphi_{5,\mathrm{II}}+\varphi_{5,\mathrm{I}}^2+\varphi_{6,\mathrm{I}}^2=0\ .
\end{aligned}\tag{26}$$

The magnetic representation corresponding to 3×6 degrees of freedom is performed by some linear combinations Ψ of the components of the spins which may easily be expressed in terms of the components of the vectors (25) written in the cyclic coordinate frame. The vectors (25) written in the cyclic coordinate frame have the form

$$\varphi_\nu=\frac{\mathbf{e}_x-i\mathbf{e}_y}{2}\varphi_\nu^+ +\frac{\mathbf{e}_x+i\mathbf{e}_y}{2}\chi_\nu^- +\varphi_\nu^z\mathbf{e}_z\ ,$$

where $\varphi_\nu^\pm=\varphi_\nu^x\pm i\varphi_\nu^y$; $\mathbf{e}_i$ are the unit vectors along the axes of the crystal coordinate frame (the Z axis coincides with the sixfold axis of the crystal lattice; see also Fig. 2). The definition of the vectors Ψ and their classification in accordance with the irreducible representations of the spatial group are given in Table II. The conventional spectroscopic notations for the irreducible representations are given in Table II along with the notations used in Ref. 37. The linear combinations Ψ which are the components of the vector $\mathbf{F}$ of the total spin of the cell are also shown in Table II. The vectors (25), corresponding to the exchange symmetry which is higher than the magnetic one, allow one to write down the exchange part of the Hamiltonian in the form

$$\mathcal{H}_{\mathrm{ex}}=J_1\varphi_1^2+J_4\varphi_4^2+J_5\varphi_{5,\mathrm{I}}\cdot\varphi_{5,\mathrm{II}}+J_6\varphi_{6,\mathrm{I}}\cdot\varphi_{6,\mathrm{II}}\ .\tag{27}$$

In this expression the quantities J are the linear combinations of the intersublattice exchange integrals $K_{\alpha\beta}$ of the form

$$\begin{aligned}
&J_1=\tfrac{1}{6}(K_{11}+2K_{12}+K_{14}+2K_{15})\ ,\\
&J_4=\tfrac{1}{6}(K_{11}+2K_{12}-K_{14}-2K_{15})\ ,\\
&J_5=\tfrac{1}{3}(-K_{11}+K_{12}+K_{14}-K_{15})\ ,\\
&J_6=\tfrac{1}{3}(K_{11}-K_{12}+K_{14}-K_{15})\ .
\end{aligned}\tag{28}$$

The conditions $K_{11}=K_{13}$ and $K_{15}=K_{16}$ have been used while obtaining expressions (28). The exact magnetic symmetry of the crystal is described by the linear combinations of the vectors Ψ. These linear combinations of the vectors Ψ describe all possible (allowed by the symmetry) contributions of a relativistic nature to the Hamiltonian. We keep only those terms which determine the activation energies of the acoustic magnons and fine details of the magnetic structure:

$$\begin{aligned}
\mathcal{H}_a=&\tfrac{1}{8}a_1^2\Psi_1^2-\tfrac{1}{8}a_2\Psi_2'^2-a_5\Psi_{5,\mathrm{V}}\Psi_{5,\mathrm{VI}}+d_2\Psi_2'\Psi_2''\\
&+\tfrac{1}{8}a_3\Psi_3'^2-\tfrac{1}{8}a_4\Psi_4^2-a_6\Psi_{6,\mathrm{V}}\Psi_{6,\mathrm{VI}}+d_3\Psi_3'\Psi_3''\ .
\end{aligned}\tag{29}$$

The magnetic ordering shown in Figs. 2 and 3 corresponds to the nonzero linear combinations of the vectors Ψ which are transformed in accordance with the T_2 and T_3 irreducible representations, respectively. In what follows we use the terms magnetically ordered phase T_2 and magnetically ordered phase T_3. The phase T_2 is realized under fulfillment of the conditions[3]

TABLE II. Classification of the components of the vectors φ in accordance with the irreducible representations of the spatial group C_{6V}^3.

Irreducible representation				
T_1; A_1	φ_1	$\Psi_1=-(\varphi_{5,\mathrm{I}}^-+\varphi_{5,\mathrm{II}}^+)$		
T_2; A_2		$\Psi_2'=\varphi_{5,\mathrm{I}}^--\varphi_{5,\mathrm{II}}^+$, $\Psi_2''=\varphi_1^z=F^z$		
T_3; B_1		$\Psi_3'=\varphi_{6,\mathrm{I}}^-+\varphi_{6,\mathrm{II}}^+$, $\Psi_3''=\varphi_4^z$		
T_4; B_2	φ_4	$\Psi_4=\varphi_{6,\mathrm{II}}^+-\varphi_{6,\mathrm{I}}^-$		
T_5; E_1	$\varphi_{5,\mathrm{I}}$	$\Psi_{5,\mathrm{I}}=\varphi_1^+=F^+$,	$\Psi_{5,\mathrm{III}}=\varphi_{6,\mathrm{II}}^-$,	$\Psi_{5,\mathrm{V}}=\varphi_{5,\mathrm{I}}^z$
	$\varphi_{5,\mathrm{II}}$	$\Psi_{5,\mathrm{II}}=\varphi_1^-=F^-$,	$\Psi_{5,\mathrm{IV}}=\varphi_{6,\mathrm{I}}^+$,	$\Psi_{5,\mathrm{VI}}=-\varphi_{5,\mathrm{II}}^z$
T_6; E_2	$\varphi_{6,\mathrm{I}}$	$\Psi_{6,\mathrm{I}}=\chi_4^+$,	$\Psi_{6,\mathrm{III}}=-\varphi_{5,\mathrm{II}}^-$,	$\Psi_{6,\mathrm{V}}=\varphi_{6,\mathrm{I}}^z$
	$\varphi_{6,\mathrm{II}}$	$\Psi_{6,\mathrm{II}}=\varphi_4^-$,	$\Psi_{6,\mathrm{IV}}=\varphi_{5,\mathrm{I}}^+$,	$\Psi_{6,\mathrm{VI}}=-\varphi_{6,\mathrm{II}}^z$

$$0<-J_5<2J_1,2J_4,J_6 \quad \text{and } a_2>a_1,-a_5>0 \ .$$

The phase T_3 corresponds to the case[3]

$$0<J_6<2J_1,2J_4,-J_5 \quad \text{and } -a_3>a_4,a_6>0 \ .$$

The magnetic symmetry of both phases allows the magnetic structures to be slightly noncoplanar. It goes without saying that the magnetic structures remain coplanar in the exchange approximation. The weak ferromagnetic moment $\overline{\Psi}_2''=\overline{F}_z$ (the overbar stands for the equilibrium value) appears in the T_2 phase and the weak antiferromagnetic moment $\overline{\Psi}_3''$ exists in the T_3 phase. The angles of canting of the equilibrium spin orientations with respect to the X,Y plane are determined by the constants d describing the Dzyaloshinskii-Moriya interactions in Hamiltonian (29). These angles are equal to

$$\sin\vartheta \simeq -d_2(J_1+\tfrac{1}{2}J_5)^{-1} \quad \text{for the phase } T_2$$

and

$$\sin\vartheta \simeq -d_3(J_4-\tfrac{1}{2}J_6)^{-1} \quad \text{for the phase } T_3 \ .$$

Thus both phases T_i ($i=2,3$) are described by the following equilibrium values:

$$\overline{\Psi}_i'=12S\cos\vartheta \approx 12S \quad \text{and } \overline{\Psi}_i''=6S\sin\vartheta \ .$$

Let us carry out the calculations of the magnon energies and coefficients of the unitary transformation. The transformation to the local coordinate systems and at the same time the classification of the oscillations of the magnetic subsystem in accordance with the types of symmetry for the T_2 phase has the form

$$\begin{aligned}
&\begin{bmatrix}\Psi_2'\\ \Psi_1\\ \Psi_2''\end{bmatrix}=\hat{p}\begin{bmatrix}\tilde{\varphi}_1^+ +\tilde{\varphi}_1^-\\ \tilde{\varphi}_1^+-\tilde{\varphi}_1^-\\ \tilde{\varphi}_1^z\end{bmatrix},\quad
\begin{bmatrix}\Psi_3'\\ \Psi_4\\ \Psi_3''\end{bmatrix}=\hat{p}\begin{bmatrix}\tilde{\varphi}_4^+ +\tilde{\varphi}_4^-\\ \tilde{\varphi}_4^+-\tilde{\varphi}_4^-\\ \tilde{\varphi}_4^z\end{bmatrix},\\
&\begin{bmatrix}\Psi_{5,\mathrm{I}}+\Psi_{5,\mathrm{III}}\\ \Psi_{5,\mathrm{I}}-\Psi_{5,\mathrm{III}}\\ \Psi_{5,\mathrm{V}}\end{bmatrix}=\hat{p}\begin{bmatrix}\tilde{\varphi}_{5,\mathrm{I}}^+ +\tilde{\varphi}_{5,\mathrm{I}}^-\\ \tilde{\varphi}_{5,\mathrm{I}}^+-\tilde{\varphi}_{5,\mathrm{I}}^-\\ \tilde{\varphi}_{5,\mathrm{I}}^z\end{bmatrix},\\
&\begin{bmatrix}\Psi_{5,\mathrm{II}}+\Psi_{5,\mathrm{IV}}\\ \Psi_{5,\mathrm{II}}-\Psi_{5,\mathrm{IV}}\\ \Psi_{5,\mathrm{VI}}\end{bmatrix}=-\hat{p}\begin{bmatrix}\tilde{\varphi}_{5,\mathrm{II}}^+ +\tilde{\varphi}_{5,\mathrm{II}}^-\\ -\tilde{\varphi}_{5,\mathrm{II}}^+ +\tilde{\varphi}_{5,\mathrm{II}}^-\\ \tilde{\varphi}_{5,\mathrm{II}}^z\end{bmatrix},\\
&\begin{bmatrix}\Psi_{6,\mathrm{I}}+\Psi_{6,\mathrm{III}}\\ \Psi_{6,\mathrm{I}}-\Psi_{6,\mathrm{III}}\\ \Psi_{6,\mathrm{V}}\end{bmatrix}=\hat{p}\begin{bmatrix}\tilde{\varphi}_{6,\mathrm{I}}^+ +\tilde{\varphi}_{6,\mathrm{I}}^-\\ \varphi_{6,\mathrm{I}}^+-\tilde{\varphi}_{6,\mathrm{I}}^-\\ \tilde{\varphi}_{6,\mathrm{I}}^z\end{bmatrix},\\
&\begin{bmatrix}\Psi_{6,\mathrm{II}}+\Psi_{6,\mathrm{IV}}\\ \Psi_{6,\mathrm{II}}-\Psi_{6,\mathrm{IV}}\\ -\Psi_{6,\mathrm{VI}}\end{bmatrix}=\hat{p}\begin{bmatrix}\tilde{\varphi}_{6,\mathrm{II}}^+ +\tilde{\varphi}_{6,\mathrm{II}}^-\\ -\tilde{\varphi}_{6,\mathrm{II}}^+ +\tilde{\varphi}_{6,\mathrm{II}}^-\\ \tilde{\varphi}_{6,\mathrm{II}}^z\end{bmatrix}.
\end{aligned} \tag{30}$$

The matrix $\hat{p}$ is connected with the matrix of the transition into the local coordinate system of the first ion:

$$\hat{p}=\begin{bmatrix}\sin\vartheta & 0 & 2\cos\vartheta\\ 0 & 1 & 0\\ -\dfrac{\cos\vartheta}{2} & 0 & \sin\vartheta\end{bmatrix}. \tag{31}$$

The orientation of the $0Z'$ axis of the local coordinate system is along the equilibrium orientation of the spin and the $0Y'$ axis is parallel to the $0Y$ axis shown in Fig. 2. The quantities $\tilde{\varphi}$ have a form similar to (25) but are constructed with the help of the linear combinations of the components of the spin operators $S_\alpha'^{\pm}$ and $S_\alpha'^z$ written in their local coordinate systems. In the approximation linear with respect to the spin-deflection operators b_α ($\alpha=1-6$) of the sublattices the quantities $\tilde{\varphi}^{\pm}$ form some generalized boson operators $\beta^\dagger$ and β of creation and annihilation in such a way that $\tilde{\varphi}_i^+=\sqrt{12S}\,\beta_i$; here i is the combined index $i=1;4;5,\mathrm{I};5,\mathrm{II};6,\mathrm{I};6,\mathrm{II}$. Clearly the operators $\beta^\dagger$ and β have the same structure as expressions (25). For example,

$$\begin{aligned}
\beta_4&=\frac{1}{\sqrt{6}}[b_1+b_2+b_3-b_4-b_5-b_6] \ ,\\
\beta_{6,\mathrm{I}}&=\frac{1}{\sqrt{6}}[b_1-\omega^*b_2-\omega b_3+b_4-\omega^*b_5-\omega b_6]=\frac{\tilde{\varphi}_{6,\mathrm{I}}^+}{\sqrt{12S}} \ ;
\end{aligned} \tag{32}$$

however,

$$\beta_{6,\mathrm{I}}^\dagger=\frac{1}{\sqrt{6}}[b_1^\dagger-\omega b_2^\dagger-\omega^*b_3^\dagger+b_4^\dagger-\omega b_5^\dagger-\omega^*b_6^\dagger] =\frac{\tilde{\varphi}_{6,\mathrm{II}}^-}{\sqrt{12S}} \ .$$

Furthermore, the ordinary commutation relations are satisfied for these operators:

$$[\beta_i,\beta_j]=[\beta_i^\dagger,\beta_j^\dagger]=0, \quad [\beta_i,\beta_j^\dagger]=\delta_{ij} \ . \tag{33}$$

Among all $\tilde{\varphi}_i^z$ we will only give the expression for $\tilde{\varphi}_1^z$ which is necessary for obtaining the part of the Hamiltonian $\mathcal{H}^{(2)}=\mathcal{H}_{\mathrm{ex}}^{(2)}+\mathcal{H}_a^{(2)}$ quadratic with respect to the operators β:

$$\tilde{\varphi}_1^z=6S-\sum_i \beta_i^\dagger\beta_i \ . \tag{34}$$

Using relations (30)–(34) one can obtain the explicit form of the Hamiltonian $\mathcal{H}^{(2)}$ in the T_2 phase:

$$\frac{\mathcal{H}^{(2)}}{3S}=\tfrac{1}{2}I_1(\beta_1+\beta_1^\dagger)^2-\tfrac{1}{2}A_1(\beta_1-\beta_1^\dagger)^2+\tfrac{1}{2}I_4(\beta_4+\beta_4^\dagger)^2-\tfrac{1}{2}i_4(\beta_4-\beta_4^\dagger)^2+(I_5+A_5+2V_5)\beta_{5,\mathrm{I}}^\dagger\beta_{5,\mathrm{I}}$$
$$+(I_5+A_5-2V_5)\beta_{5,\mathrm{II}}^\dagger\beta_{5,\mathrm{II}}-(I_5-A_5)(\beta_{5,\mathrm{I}}^\dagger\beta_{5,\mathrm{II}}^\dagger+\beta_{5,\mathrm{I}}\beta_{5,\mathrm{II}})+(I_6+i_6+2V_6)\beta_{6,\mathrm{I}}^\dagger\beta_{6,\mathrm{I}}$$
$$+(I_6+i_6-2V_6)\beta_{6,\mathrm{II}}^\dagger\beta_{6,\mathrm{II}}-(I_6-i_6)(\beta_{6,\mathrm{I}}^\dagger\beta_{6,\mathrm{II}}^\dagger+\beta_{6,\mathrm{I}}\beta_{6,\mathrm{II}})\ . \qquad (35)$$

The following notations are introduced here:

$$\begin{aligned}
&I_1=(2J_1+J_5+a_2)\cos 2\vartheta-4d_2\sin 2\vartheta\ ,\\
&I_4=2J_4\cos^2\vartheta+(J_6+a_3)\sin^2\vartheta-2d_3\sin 2\vartheta+\Delta\ ,\\
&I_5=J_1+\tfrac{1}{2}J_6+\Delta,\quad I_6=J_4-\tfrac{1}{2}J_5+\Delta\ ,\\
&A_1=a_2-a_1-2d_2\tan\vartheta,\quad \Delta=J_5+a_2-2d_2\tan\vartheta\ ,\\
&A_5=a_2-a_5+(J_1+\tfrac{1}{2}J_6+J_5+a_5)\sin^2\vartheta-2d_2\tan\vartheta\ ,\\
&i_4=J_6+a_4+\Delta\ ,\\
&i_6=(J_6+a_6)\cos^2\vartheta+(J_4-\tfrac{1}{2}J_5)\sin^2\vartheta+\Delta\ ,\\
&V_5=(J_1-\tfrac{1}{2}J_6)\sin\vartheta,\quad V_6=(J_4+\tfrac{1}{2}J_5)\sin\vartheta\ .
\end{aligned} \qquad (36)$$

The sets of the constants of the Hamiltonian in which the integral of the intralayer exchange K_{12} will remain after the transition to the exchange approximation are denoted by I. The quantities i will only contain the integrals K_{14} and K_{15} of the interlayer exchange in the exchange approximation. All the terms entering the quantities A have the order of magnitude of the anisotropy because in magnets for which the exchange approximation $J \gg d \gg a$ is valid the estimation $a \sim d^2J^{-1}$ is true as a rule. The quantities V_5 and V_6 are of the order of the constants d describing the Dzyaloshinskii-Moriya interactions.

The diagonalization of Hamiltonian (35) gives the following expressions for the magnon energies:

$$\begin{aligned}
&\varepsilon_1^{(A)}=6S\sqrt{I_1A_1},\quad \varepsilon_{5,\pm}^{(A)}=6S[\sqrt{I_5A_5}\pm V_5]\ ,\\
&\varepsilon_4^{(E)}=6S\sqrt{I_4i_4},\quad \varepsilon_{6,\pm}^{(E)}=6S[\sqrt{I_6i_6}\pm V_6]\ .
\end{aligned} \qquad (37)$$

In these expressions the energy superscript signifies that this frequency corresponds to the acoustic or exchange magnons. For investigation of the one-magnon light scattering one has to obtain the parts of the combinations Ψ linear with respect to the operators $\xi_\nu^\dagger$ and ξ_ν of creation and annihilation of the magnons of the νth branch. The above-mentioned parts of the operators Ψ may be obtained as a result of the calculation of the u-v coefficients of the unitary transformation from the operators $\beta^\dagger$ and β to the operators $\xi_\nu^\dagger$ and ξ_ν. Finally the expressions (30) may be rewritten as follows:

$$\begin{aligned}
&\begin{bmatrix}\Psi_2'\\ \Psi_1\\ \Psi_2''\end{bmatrix}=\sqrt{12S}\hat{p}\begin{bmatrix}(A_1/I_1)^{1/4}(\xi_1+\xi_1^\dagger)\\ (I_1/A_1)^{1/4}(\xi_1-\xi_1^\dagger)\\ 0\end{bmatrix}\ ,\\
&\begin{bmatrix}\Psi_3'\\ \Psi_4\\ \Psi_3''\end{bmatrix}=\sqrt{12S}\hat{p}\begin{bmatrix}(i_4/I_4)^{1/4}(\xi_4+\xi_4^\dagger)\\ (I_4/i_4)^{1/4}(\xi_4-\xi_4^\dagger)\\ 0\end{bmatrix}\ ,\\
&\begin{bmatrix}\Psi_{5,\mathrm{I}}+\Psi_{5,\mathrm{III}}\\ \Psi_{5,\mathrm{I}}-\Psi_{5,\mathrm{III}}\\ \Psi_{5,\mathrm{V}}\end{bmatrix}=\sqrt{12S}\hat{p}\begin{bmatrix}(I_5/A_5)^{1/4}(\xi_{5,+}+\xi_{5,-}^\dagger)\\ (A_5/I_5)^{1/4}(\xi_{5,+}-\xi_{5,-}^\dagger)\\ 0\end{bmatrix}\ ,\\
&\begin{bmatrix}\Psi_{6,\mathrm{I}}+\Psi_{6,\mathrm{III}}\\ \Psi_{6,\mathrm{I}}-\Psi_{6,\mathrm{III}}\\ \Psi_{6,\mathrm{V}}\end{bmatrix}=\sqrt{12S}\hat{p}\begin{bmatrix}(I_6/i_6)^{1/4}(\xi_{6,+}+\xi_{6,-}^\dagger)\\ (i_6/I_6)^{1/4}(\xi_{6,+}-\xi_{6,-}^\dagger)\\ 0\end{bmatrix}\ .
\end{aligned} \qquad (38)$$

The two columns which are Hermitian conjugate to the last two are not written here. The zero value in the bottom lines of the columns in the right-hand side of expressions (38) means that the corresponding contribution is quadratic with respect to the creation and annihilation operators and has to be neglected for the one-magnon light scattering.

Let us dwell on the formulas (37) and (38). The unitary point subgroup corresponding to the phase T_2 is $C_6(6)$, which has no two-dimensional representations. Due to this circumstance the degeneration of the magnons at $\mathbf{k}=\mathbf{0}$ is absent. The degeneration appears only at $d_2=0$. Similarly to the case of collinear magnets the activation energies of the acoustic magnons are of the order of $\varepsilon_A \sim \sqrt{Ja}$ where the quantity J contains the intralayer exchange integral. The splitting of the energies of the acoustic magnons $\varepsilon_{5,\pm}^{(A)}$ may be important because $\sqrt{Ja} \sim d$. The activation energies of the exchange magnons are determined by the interlayer exchange integrals. Indeed, using (28), one can obtain in the exchange approximation

$$\varepsilon_4^{(E)}=\sqrt{2}\varepsilon_{6,\pm}^{(E)}=6S\sqrt{\tfrac{1}{3}(K_{12}-K_{15})(K_{14}-K_{15})}\ .$$

The integrals of the interlayer exchange are often considerably smaller than the integrals of the intralayer exchange. A similar situation taking place in the four-sublattice antiferromagnet La_2CuO_4 leads to the fact that the energy of the exchange magnons falls within the region of the energies of the acoustic magnons.

It follows from formulas (37) that during the oscillations corresponding to the acoustic magnon modes the largest [because of the presence of the factor $(J/A)^{1/4}$] deviations of the spins from the equilibrium orientations correspond to the rotation of the magnetic structure as a whole. That is, the spin triangules in the neighboring layers precess in phase. The rotations of the magnetic structure inside the X,Y plane (in the plane of the layer) correspond to the acoustic mode with the energy $\varepsilon_1^{(A)}$. The rotations during which the magnetic structure leaves the X,Y plane correspond to the acoustic modes with energies $\varepsilon_{5,\pm}^{(A)}$.

Let us consider the spin-dependent part of the dielectric permittivity of $R\mathrm{MnO}_3$ which we will write using the coordinates $\zeta=x+iy$ and $\eta=x-iy$ in the X,Y plane. For the sake of simplicity we will keep only those terms quadratic with respect to the spins which have the exchange nature

$$\begin{aligned}
&\Delta\varepsilon_{\zeta\eta}=i\lambda_1\Psi_2'+i\lambda_2\Psi_2''+P_1\varphi_1^2+P_2\varphi_4^2+P_3\varphi_{5,\mathrm{I}}\cdot\varphi_{5,\mathrm{II}}\ ,\\
&\Delta\varepsilon_{\zeta\zeta}=P_4\varphi_1\cdot\varphi_{6,\mathrm{I}}+P_5\varphi_4\cdot\varphi_{5,\mathrm{I}}+P_6\varphi_{5,\mathrm{II}}^2\ ,\\
&\Delta\varepsilon_{zz}=P_7\varphi_1^2+P_8\varphi_4^2+P_9\varphi_{5,\mathrm{I}}\cdot\varphi_{5,\mathrm{II}}\ ,\\
&\Delta\varepsilon_{\eta z}=i\lambda_3\Psi_{5,\mathrm{I}}+i\lambda_4\Psi_{5,\mathrm{III}}+i\lambda_5\Psi_{5,\mathrm{V}}\\
&\qquad\quad+P_{10}\varphi_1\cdot\varphi_{5,\mathrm{I}}+P_{11}\varphi_4\cdot\varphi_{6,\mathrm{I}}\ ,\\
&\Delta\varepsilon_{\eta\zeta}=(\Delta\varepsilon_{\zeta\eta})^*,\quad \Delta\varepsilon_{\eta\eta}=(\Delta\varepsilon_{\zeta\zeta})^*,\quad \Delta\varepsilon_{\zeta z}=(\Delta\varepsilon_{\eta z})^*\ .
\end{aligned}\tag{39}$$

The orthogonality relations (26) have been used while writing expressions (39). The presence of the contributions linear not only with respect to the components of the ferromagnetism vector but also with respect to the components of the antiferromagnetism vector is a well-known phenomenon and in weak ferromagnets that has been studied in sufficient detail, for example, in orthoferrites.[20,35] Such terms have to appear in all magnets in which the components of the antiferromagnetism vectors are transformed in accordance with the same irreducible representations as the components of the ferromagnetism vector. In the case of the light scattering from magnons such contributions have been considered by Moriya.[33]

For the sake of convenience the Cartesian indices of the electric field of the incident and scattered light are used in the quantities $\Delta\varepsilon_{ij}$ and further in the scattering tensor $a_{ij}^{(\nu)}$. Let us recall that in terms of the cyclic coordinates the electric-field energy is written in the form $\Delta\varepsilon_{i^*j^*}E_iE_j$, for example, $\Delta\varepsilon_{\eta z}E_\eta E_z$.

The form of the tensors of scattering and the explicit form of their components one can obtain with the help of formula (1) and the substitution of expressions (38) in (39). As a result we obtained the following.

(a) For the acoustic mode A_1,

$$a_{ij}^{(A_1)}=\begin{vmatrix} 0 & a_{\zeta\eta} & 0\\ a_{\eta\zeta} & 0 & 0\\ 0 & 0 & a_{zz}\end{vmatrix}\ .\tag{40}$$

Here

$$a_{\zeta\eta}=\sqrt{12S}\left[\frac{A_1}{I_1}\right]^{1/4}\left\{i\lambda_1\sin\vartheta-\tfrac{1}{2}\lambda_2\cos\vartheta-\frac{3S}{2}(2P_1+P_3)\sin2\vartheta\right\},$$

$$a_{zz}=-\sqrt{12S}\left[\frac{A_1}{I_1}\right]^{1/4}\frac{3S}{2}(2P_7+P_9)\sin2\vartheta\ .$$

Let us recall that the quantities P are symmetric whereas the quantities λ are antisymmetric tensors with respect to the Cartesian indices of the incident and scattered light. In addition, the quantities P and λ are real in the framework of the present consideration. It follows from (40) that the contribution of the exchange mechanism of light scattering from the acoustic magnons contains two small parameters, namely, the factor $(A/J)^{1/4}$ and $\sin\vartheta$ which determines the weak noncoplanarity of the magnetic structure.

The Stokes–anti-Stokes asymmetry does not appear in the approximation used while obtaining the expressions for $\Delta\varepsilon$. However, in case we take into account the terms of a relativistic nature in $\Delta\varepsilon_{\zeta\eta}$ quadratic with respect to the spins, this asymmetry appears. For example, a term of the form

$$R\Psi_1\Psi_2'\sim\sqrt{12S}R(I_1/A_1)^{1/4}(\xi_1-\xi_1^\dagger)$$

which is exchange enhanced will enter the tensors of the Stokes and anti-Stokes scattering with different signs.

The given example explains in general the mechanism of the appearance of the Stokes–anti-Stokes asymmetry. Such an asymmetry always appears when the parts of $\Delta\varepsilon$ linear with respect to ξ and $\xi^\dagger$ operators have the form $(\xi_1-\xi_1^\dagger)$. In particular, these terms were absent in the expressions for $\Delta\varepsilon$ in the case of UO_2.

(b) For the acoustic modes $A_{5,\pm}$ the scattering tensors have the form

$$\begin{aligned}
a_{ij}^{(A_{5,+})}&=\begin{vmatrix} 0 & 0 & a_{\zeta z}\\ 0 & 0 & 0\\ (a_{\zeta z})^* & 0 & 0\end{vmatrix}\ ,\\
a_{ij}^{(A_{5,-})}&=\begin{vmatrix} 0 & 0 & 0\\ 0 & 0 & a_{\eta z}\\ 0 & (a_{\eta z})^* & 0\end{vmatrix}\ .
\end{aligned}\tag{41}$$

Here

$$a_{x\pm iy,z}^{(A_{5,\pm})}=\sqrt{3S}\left\{\mp[(i\lambda_3+i\lambda_4\pm3SP_{10}\cos\vartheta)\sin\vartheta-i\lambda_5\cos\vartheta]\left[\frac{I_5}{A_5}\right]^{1/4}-(i\lambda_3-i\lambda_4\mp3SP_{10}\cos\vartheta)\left[\frac{A_5}{I_5}\right]^{1/4}\right\}.\tag{42}$$

In these expressions, similar to the case of light scattering from the acoustic magnons in exchange-collinear orthoferrites,[30] there exists the effect of the exchange enhancement of the antisymmetric part of the scattering tensor, which is proportional to λ_5 in the present case. Let us recall that this quantity determines the contribution of the antiferromagnetism vector $\Psi_{5,\mathrm{V}}$ in linear magneto-optic effects. Using the terminology of Ref. 20 one can say that these linear terms, which are proportional to the antiferromagnetism vectors, describe the out-of-phase scattering. However, comparing formula (42) and the expression for $a_{\zeta\eta}$ in (40), one can see that the contribution of the above-mentioned terms is not necessarily exchange enhanced [that is, containing the factor $(J/A)^{1/4}$], in contrast with the example given in Ref. 20. The contribution of the magneto-optic term of the exchange nature P_{10} is weakened due to the presence of the factor $\sin\vartheta$ in spite of the presence of the factor $(J/A)^{1/4}$. Nevertheless, the appearance of this term together with the ordinary Faraday summand $i\lambda_3$ in the terms proportional to $(A/J)^{1/4}$ can testify that the value of the symmetric part of the off-diagonal components of the tensors $a_{ij}^{(A_{5,\pm})}$ may be considerable. The quantity P_{10} expressed through the spin-dependent exchange part of the polarizability of the pair of ions belonging to α and β sublattices $\pi^{ij}_{\alpha\beta}=\frac{1}{3}\mathrm{Tr}(\pi^{ijll}_{\alpha\beta})$ has the form

$$P_{10}=\frac{1}{2}\left[\pi^{xz}_{12}-\sqrt{3}\pi^{yz}_{12}+\pi^{xz}_{15}-\frac{1}{\sqrt{3}}\pi^{yz}_{15}\right] .$$

In connection with the aforesaid it is interesting to carry out measurements of the tensor of scattering from the $A_{5,\pm}$ acoustic magnons in the presence of an external magnetic field oriented along the Z axis (the sixfold axis of the crystal). The quantity $\sin\vartheta$ will increase linearly with increasing field in this case. The contribution to the scattering tensor proportional to P_{10} and at the same time proportional to $(J/A)^{1/4}\sin\vartheta$ will also increase.

(c) For the exchange magnons $E_{6,\pm}$ the scattering tensors are as follows:

$$a_{ij}^{(E_{6,+})}=\begin{bmatrix}0&0&0\\0&a_{\eta\eta}&0\\0&0&0\end{bmatrix} ,$$
$$a_{ij}^{(E_{6,-})}=\begin{bmatrix}a_{\zeta\zeta}&0&0\\0&0&0\\0&0&0\end{bmatrix} . \tag{43}$$

Here

$$a^{(E_{6,\mp})}_{x\pm iy,x\pm iy}=(3S)^{3/2}\cos\vartheta \times\left\{(-2P_4+P_5+P_6)\sin\vartheta\left[\frac{I_6}{i_6}\right]^{1/4} \mp(P_5-P_6)\left[\frac{i_6}{I_6}\right]^{1/4}\right\} . \tag{44}$$

As follows from this expression, the scattering from the exchange magnons $E_{6,\pm}$ is determined by the exchange magneto-optic constants only. The main contribution which does not contain any small parameters is given by the term proportional to (P_5-P_6). This term may be expressed through the quantities $\pi^{ij}_{\alpha\beta}$ as follows:

$$(P_5-P_6)=\tfrac{1}{6}\{\pi^{xx}_{11}-\pi^{yy}_{11}+2(\pi^{xx}_{12}-\pi^{yy}_{12})-\pi^{xx}_{14}+\pi^{yy}_{14} -2(\pi^{xx}_{15}-\pi^{yy}_{15}) +2\sqrt{3}(\pi^{xy}_{12}+\pi^{yx}_{12}-\pi^{xy}_{15}-\pi^{yx}_{15})\} .$$

Note that in the case of the exchange-collinear antiferromagnet La_2CuO_4 the contribution of the exchange mechanism into the tensor of scattering from the exchange magnons was exchange weakened by the small factor determining the small noncollinearity of the magnetic structure.[31]

Thus the scattering from the exchange magnons in exchange-noncollinear magnets of the RMnO$_3$ type exists even in the exchange approximation. The exchange mode E_4 is a silent mode, that is, in the absence of an external magnetic field this mode is manifested neither in infrared spectra nor in Raman ones.

Let us take into account the nonanalytic part of the dipole-dipole interaction for the case of RMnO$_3$-type magnets. In the general case without utilizing the approximation of the isotropic g factor the contribution of this part of the dipole-dipole interaction to the Hamiltonian has the form

$$\mathcal{H}_{d\text{-}d}=\frac{2\pi}{v_0}\mu_B^2\frac{1}{|\mathbf{q}|^2}\left[\sum_{ij,\alpha}g^{(\alpha)}_{ij}q_iS_{j\alpha}\right]^2 . \tag{45}$$

The positional symmetry of the local surrounding of the manganese ions leads to the following form of the tensor of the g factor, for example, of the first ion:

$$g^{(1)}_{ij}=\begin{bmatrix}g^{(1)}_{xx}&0&g^{(1)}_{xz}\\0&g^{(1)}_{yy}&0\\g^{(1)}_{zx}&0&g^{(1)}_{zz}\end{bmatrix} . \tag{46}$$

Using this form of the tensor and also the symmetry relations between the tensors of the g factor for different Mn ions, the sum entering the expression for $\mathcal{H}_{d\text{-}d}$ may be rewritten in terms of the Ψ operators as

$$\sum_{ij,\alpha}g^{(\alpha)}_{ij}q_iS_{j\alpha}=\tfrac{1}{4}q_-[g^{(1)}_{xx}(\Psi_{5,\mathrm{I}}+\Psi_{5,\mathrm{III}})+g^{(1)}_{yy}(\Psi_{5,\mathrm{I}}-\Psi_{5,\mathrm{III}})+2g^{(1)}_{xz}\Psi_{5,\mathrm{V}}] +\tfrac{1}{4}q_+[g^{(1)}_{xx}(\Psi_{5,\mathrm{II}}+\Psi_{5,\mathrm{IV}})+g^{(1)}_{yy}(\Psi_{5,\mathrm{II}}-\Psi_{5,\mathrm{IV}})+2g^{(1)}_{xz}\Psi_{5,\mathrm{VI}}]+q_z[\tfrac{1}{2}g^{(1)}_{zx}\Psi'_2+g^{(1)}_{zz}\Psi''_2] . \tag{47}$$

In correspondence with the symmetry, all the operators Ψ which are transformed like the components of the total spin of the crystal (see Table II) entered the sum in (47).

For a better understanding of what follows, let us note that the response of the crystal to the external magnetic field $\mathbf{H}$ is in general determined by the quantity

$$M_i = -\frac{\delta \mathcal{H}}{\delta H_i} = \mu_B \sum_{j,\alpha} g_{ij}^{(\alpha)} S_{j,\alpha} ,$$

which one can determine as the magnetic moment of the magnetic cell of the crystal. If the g factors of the ions are anisotropic the tensors $g_{ij}^{(\alpha)}$ contain the off-diagonal components. In this case the components of the total spin of the cell (the components of the ferromagnetism vector) and the components of the antiferromagnetism vector which are transformed in the same manner during the symmetry operations give a contribution to the quantity M. For example, in the case of $R\mathrm{MnO}_3$ we have

$$M_z = \mu_B (\tfrac{1}{2} g_{zx}^{(1)} \Psi_2' + g_{zz}^{(1)} \Psi_2'') ,$$

where Ψ_2'' is the component of the ferromagnetism vector. In the case of UO_2 the components of the total spin only entered the expression for $\mathbf{M}$.

The definition of $\mathbf{M}$ given above allows us to write the nonanalytic part of the dipole-dipole interaction Hamiltonian for a magnet of any symmetry in the form

$$\mathcal{H}_{d\text{-}d} = \frac{2\pi}{v_0} \frac{(\mathbf{q}\cdot\mathbf{M})^2}{|\mathbf{q}|^2} . \tag{48}$$

That is why in what follows, when speaking about the longitudinal or transverse magnons, one has to understand that the case in point is about the oscillations of the vector of the total magnetic moment of the cell which are longitudinal or transverse with respect to the direction of the magnon propagation, rather than the oscillations of the vector of the total spin.

Using relations (45), (47), and (38), one can obtain the part of $\mathcal{H}_{d\text{-}d}$ for the T_2 magnetic phase quadratic with respect to the operators of creation and annihilation of the magnons. As a result one can get

$$\mathcal{H}_{d\text{-}d} = \{ g_+ (f_+ \xi_2^\dagger + f_- \xi_2) - g_- (f_+ \xi_3 + f_- \xi_3^\dagger) + g_z f_z (\xi_1^\dagger + \xi_1) \}^2 . \tag{49}$$

The following notations are used here:

$$g_\pm = \left[\frac{3S\pi}{2v_0} \right]^{1/2} \mu_B \left\{ \pm (g_x^{(1)} \sin\vartheta - g_{xz}^{(1)} \cos\vartheta) \left[\frac{I_5}{A_5} \right]^{1/4} + g_{yy}^{(1)} \left[\frac{A_5}{I_5} \right]^{1/4} \right\} ,$$

$$g_z = \left[\frac{6S\pi}{v_0} \right]^{1/2} \mu_B (g_{zx}^{(1)} \sin\vartheta - g_{zz}^{(1)} \cos\vartheta) \left[\frac{A_1}{I_1} \right]^{1/4} ,$$

$$\mathbf{f} = \frac{\mathbf{q}}{|\mathbf{q}|} .$$

It is necessary to note the presence of the exchange-enhanced term in the quantities $g_\pm$. Notice also that due to Moriya's paper[43] the quantities g_{xz} and $\sin\vartheta$ have the same microscopic nature. Because of this if one neglects the canting angle ϑ one has at the same time to put the value g_{xz} equal to zero.

Since the frequencies of all magnetodipole-active acoustic magnons are nondegenerate in a given magnetically ordered phase T_2 taking into account the dipole-dipole interaction will not lead to the appearance of pure longitudinal or transverse acoustic magnons. The only exceptions are the cases when $\mathbf{q} \| 0Z$ and $\mathbf{q} \perp 0Z$. In the first case by symmetry of the alignment of the vector $\mathbf{q}$ the oscillations of the components of the vector of the magnetic moment are longitudinal for the $\varepsilon_1^{(A)}$ magnon and transverse for the $\varepsilon_{5,\pm}^{(A)}$ magnons. In the second case $\mathbf{q} \perp 0Z$ the oscillations of the M_z component are transverse for the $\varepsilon_1^{(A)}$ magnon. As to the $\varepsilon_{5,\pm}^{(A)}$ magnons their polarization will be oblique as will be clear from what follows.

For the other directions of the vector $\mathbf{q}$ the polarization of all the acoustic magnons will be oblique due to the mixing of states caused by the dipole-dipole interaction. A similar situation takes place for the oscillations of the electric-dipole moment of the cell during the propagation of polar optic phonons in crystals belonging to the lowest-symmetry crystal classes.

Consider the mixing of states and the form of the scattering tensors in the simplest case $\mathbf{q} \perp 0Z$. Since only the two first terms inside the curly brackets on the left-hand side of (49) will remain in this case the energy $\varepsilon_1^{(A)}$ is unchanged. The energies of the two other acoustic magnons obtained after diagonalization of the Hamiltonian $\mathcal{H}^{(2)} + \mathcal{H}_{d\text{-}d}^{(2)}$ turn out to be independent of the orientation of the vector $\mathbf{q}$ in the X, Y plane. These energies have the form

$$\Omega_\pm^2 = \tfrac{1}{2} \{ \varepsilon_2^2 + \varepsilon_3^2 + 4g_+^2 \varepsilon_2 + 4g_-^2 \varepsilon_3 \pm [C^2 + T^2]^{1/2} \} .$$

The following notations are introduced here:

$$C = \varepsilon_2^2 - \varepsilon_3^2 + 4g_+^2 \varepsilon_2 - 4g_-^2 \varepsilon_3 ,$$

$$T = 4g_+ g_- \sqrt{\varepsilon_2 \varepsilon_3}, \quad \varepsilon_2 = \varepsilon_{5,+}^{(A)}, \quad \varepsilon_3 = \varepsilon_{5,-}^{(A)} .$$

The intensity of the scattering has the simplest form in the case when the scattering plane coincides with the XY plane. The mutual orientations of the wave vectors of the incident k_i and scattered k_s light are shown in Fig. 4. The components of the vector $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_s$ may be easily expressed through the angle ϕ and the scattering angle ψ if we assume that $|\mathbf{k}_i| = |\mathbf{k}_s|$ because of the inequality $\omega_i, \omega_s \gg \Omega_\pm$. Taking into account the dipole-dipole interaction leads to the result that the components of the tensors of scattering from the mixed states of the acoustic magnons depend on the scattering angle,

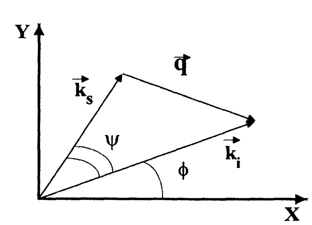

FIG. 4. The mutual orientations of the wave vectors of incident and scattered light.

$$a_{\eta z}^{(\Omega_\pm)}=\frac{1}{2}\left\{(a_{\zeta z})^*\frac{\varepsilon_2-\Omega_\pm}{\varepsilon_2}t_{2,\pm}+a_{\eta z}\frac{\varepsilon_3+\Omega_\pm}{\varepsilon_3}t_{3,\pm}\right\}\times\exp i\left[\phi+\frac{\psi-\pi}{2}\right],$$
$$a_{\zeta z}^{(\Omega_\pm)}=\frac{1}{2}\left\{a_{\zeta z}\frac{\varepsilon_2+\Omega_\pm}{\varepsilon_2}t_{2,\pm}+(a_{\eta z})^*\frac{\varepsilon_3-\Omega_\pm}{\varepsilon_3}t_{3,\pm}\right\}\times\exp\left[-i\left[\phi+\frac{\psi-\pi}{2}\right]\right]. \quad (50)$$

The quantities $a_{z\eta}^{(\Omega_\pm)}$ and $a_{z\zeta}^{(\Omega_\pm)}$ may be obtained from (50) by the permutation of the complex-conjugation sign over components $a_{\zeta z}$ and $a_{\eta z}$, respectively. The coefficients t_2 and t_3 of the new unitary transformation have the form

$$t_{2,\pm}=\pm\left[\frac{\varepsilon_2}{2\Omega_\pm}\left[1\pm\frac{C}{\sqrt{C^2+4T^2}}\right]\right]^{1/2},$$
$$t_{3,\pm}=-\left[\frac{\varepsilon_3}{2\Omega_\pm}\left[1\mp\frac{C}{\sqrt{C^2+4T^2)}}\right]\right]^{1/2}.$$

The relationships (50) have to be taken into account during the determination of the symmetric and antisymmetric parts of the tensor of the scattering from the acoustic magnons.

It is of interest to determine the dipole-dipole contribution to the energy of the acoustic magnons. To do this one has to compare the energies $\Omega_\pm$ of the acoustic magnons in the case when $\mathbf{q}\perp 0Z$ and the energies ε_2 and ε_3 which will be observed at $\mathbf{q}\|0Z$.

Let us consider the magnetically ordered phase T_3. The transition to the local coordinate systems for the functions Ψ is described by the formulas (30) in which one has to change

$$\Psi_2\Longrightarrow\Psi_3,\quad \Psi_{5,\mathrm{I}}+\Psi_{5,\mathrm{III}}\Longrightarrow\Psi_{6,\mathrm{I}}+\Psi_{6,\mathrm{III}},$$
$$\Psi_{5,\mathrm{V}}\Longrightarrow\Psi_{6,\mathrm{V}},$$
$$\Psi_{5,\mathrm{II}}+\Psi_{5,\mathrm{IV}}\Longrightarrow\Psi_{6,\mathrm{II}}+\Psi_{6,\mathrm{IV}},\quad \Psi_{5,\mathrm{VI}}\Longrightarrow\Psi_{6,\mathrm{VI}}.$$

The form of the $\hat{p}$ matrix remains the same. Carrying out the same calculations as in the case of the T_2 magnetic phase, one obtains the energies of the magnons in the magnetically ordered phase T_3:

$$\varepsilon_1^{(E)}=6S\sqrt{\tilde{I}_1\tilde{i}_1},\quad \varepsilon_4^{(A)}=6S\sqrt{\tilde{I}_4\tilde{i}_4},$$
$$\varepsilon_\pm=3S\{\tilde{I}_5\tilde{A}_5+\tilde{I}_6\tilde{i}_6+2V_5V_6 \pm[(\tilde{I}_5\tilde{A}_5-\tilde{I}_6\tilde{i}_6)^2 +4(V_5\tilde{A}_5+V_6\tilde{i}_6)(V_5\tilde{I}_6+V_6\tilde{I}_5)]^{1/2}\}^{1/2}. \quad (51)$$

The quantities $\tilde{I}$, $\tilde{i}$, and $\tilde{A}$ may be obtained from the formulas for I, i, and A for the T_2 phase by means of the following changes:

$$J_1\Longrightarrow J_4,\quad J_5\Longrightarrow -J_6,\quad d_2\Longrightarrow d_3,$$
$$a_1\Longrightarrow -a_4,\quad a_2\Longrightarrow -a_3,\quad a_5\Longrightarrow a_6.$$

Then one can obtain that

$$I_1\Longrightarrow\tilde{I}_1,\quad I_4\Longrightarrow\tilde{I}_4,\quad I_5\Longrightarrow\tilde{I}_6,\quad I_6\Longrightarrow\tilde{I}_5,$$
$$i_4\Longrightarrow\tilde{i}_1,\quad i_6\Longrightarrow\tilde{i}_6,\quad A_1\Longrightarrow\tilde{A}_4,\quad A_5\Longrightarrow\tilde{A}_5.$$

The quantities V_5 and V_6 are determined in the same way as in the phase T_2.

The energies of the two acoustic magnons ε_+ and two exchange magnons ε_- from formulas (51) are 2×2 degenerate from the fact that they belong to the two-dimensional representation of the group $C_{3V}(3mm)$ which in turn is the unitary point subgroup of the symmetry group of the T_3 magnetic phase. The states of these magnons are described by the linear combinations of Ψ_5 and Ψ_6 and belong to the same representation of the unitary subgroup. By virtue of this fact there is mixing of these states. The magnitude of this mixing is determined by the weak noncollinearity of the magnetic structure, that is, by $\sin\vartheta$. The account of this mixing is not of fundamental importance for the consideration of the light scattering and will not be taken into account in all further calculations. For our purposes it is enough to keep $\sin\vartheta$ in the matrix $\hat{p}$, putting it to be equal to zero during the calculation of the u-v coefficients and in expressions (51) for the magnon energies $\varepsilon_\pm$.

The account of the mixing of states for exchange and acoustic magnons is of fundamental importance during the investigation of the magnetic resonance of exchange magnons. The situation which appears in this case is similar to the one which occurs in exchange-collinear magnets.[44] The exchange modes manifest themselves in magnetic resonance only owing to those relativistic terms in the Hamiltonian of the magnet which lead or can lead to a weak disruption of the initial type of exchange ordering.

Taking into account all the abovesaid one can represent the parts of the operators Ψ linear with respect to the operators $\xi_\nu^\dagger$ and ξ_ν of creation and annihilation of magnons. For the case of the magnetic phase T_3 they have the form

$$\begin{bmatrix}\Psi_3'\\ \Psi_1\\ \Psi_3''\end{bmatrix}=\sqrt{12S}\hat{p}\begin{bmatrix}(\tilde{i}_1/\tilde{I}_1)^{1/4}(\xi_1+\xi_1^\dagger)\\ (\tilde{I}_1/\tilde{i}_1)^{1/4}(\xi_1-\xi_1^\dagger)\\ 0\end{bmatrix},$$
$$\begin{bmatrix}\Psi_2'\\ \Psi_4\\ \Psi_2''\end{bmatrix}=\sqrt{12S}\hat{p}\begin{bmatrix}(\tilde{A}_4/\tilde{I}_4)^{1/4}(\xi_4+\xi_4^\dagger)\\ (\tilde{I}_4/\tilde{A}_4)^{1/4}(\xi_4-\xi_4^\dagger)\\ 0\end{bmatrix}, \quad (52)$$
$$\begin{bmatrix}\Psi_{6,\mathrm{I}}+\Psi_{6,\mathrm{III}}\\ \Psi_{5,\mathrm{I}}-\Psi_{5,\mathrm{III}}\\ \Psi_{6,\mathrm{V}}\end{bmatrix}=\sqrt{6S}\hat{p}\begin{bmatrix}(\tilde{I}_5/\tilde{A}_5)^{1/4}(\xi_{5,\mathrm{I}}-\xi_{5,\mathrm{II}}+\xi_{5,\mathrm{I}}^\dagger+\xi_{5,\mathrm{II}}^\dagger)\\ (\tilde{A}_5/\tilde{I}_5)^{1/4}(\xi_{5,\mathrm{I}}-\xi_{5,\mathrm{II}}-\xi_{5,\mathrm{I}}^\dagger-\xi_{5,\mathrm{II}}^\dagger)\\ 0\end{bmatrix},$$

$$\begin{bmatrix} \Psi_{5,\mathrm{I}}+\Psi_{5,\mathrm{III}} \\ \Psi_{6,\mathrm{I}}-\Psi_{6,\mathrm{III}} \\ \Psi_{5,\mathrm{V}} \end{bmatrix} = \sqrt{6S}\hat{p} \begin{bmatrix} (\tilde{I}_6/\tilde{i}_6)^{1/4}(\xi_{6,\mathrm{I}}-\xi_{6,\mathrm{II}}+\xi^\dagger_{6,\mathrm{I}}+\xi^\dagger_{6,\mathrm{II}} \\ (\tilde{i}_6/\tilde{I}_6)^{1/4}(\xi_{6,\mathrm{I}}-\xi_{6,\mathrm{II}}-\xi_{6,\mathrm{I}}-\xi^\dagger_{6,\mathrm{II}}) \\ 0 \end{bmatrix} .$$

The two columns Hermitian conjugated to the last two columns of (52) have to be added to the above expressions. The roman digit in the subscripts of the operators ξ_5 and ξ_6 numbers the twice-degenerate states. In our approximations the energies of these states are equal to $\varepsilon_5^{(A)}=\sqrt{\tilde{I}_5\tilde{A}_5}$ for the acoustic magnons and $\varepsilon_6^{(E)}=\sqrt{\tilde{I}_6\tilde{i}_6}$ for the exchange magnons.

Consider now the tensors of scattering for the T_3 magnetic phase. Using relationships (52) and (39) we get the following.

(a) For the $\varepsilon_1^{(E)}$ exchange magnon

$$a_{ij}^{(E_1)} = \begin{bmatrix} 0 & a_{\xi\eta}^{(1)} & 0 \\ a_{\xi\eta}^{(1)} & 0 & 0 \\ 0 & 0 & a_{zz} \end{bmatrix} , \tag{53}$$

where

$$a_{\xi\eta}^{(1)} = -2(3S)^{3/2}P_2\sin2\vartheta \left[\frac{\tilde{i}_1}{\tilde{I}_1}\right]^{1/4} ,$$

$$a_{zz}=a_{\xi\eta}^{(1)}(P_2 \Longrightarrow P_8) .$$

(b) For the $\varepsilon_4^{(A)}$ acoustic magnon

$$a_{ij}^{(A_4)} = \begin{bmatrix} 0 & a_{\xi\eta}^{(4)} & 0 \\ (a_{\xi\eta}^{(4)})^* & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} . \tag{54}$$

Here

$$a_{\xi\eta}^{(4)}=i\sqrt{12S}\left[\frac{\tilde{A}_4}{\tilde{I}_4}\right]^{1/4}[\lambda_1\sin\vartheta-\tfrac{1}{2}\lambda_2\cos\vartheta] .$$

(c) For the acoustic and exchange magnons which have the energies $\varepsilon_5^{(A)}$ and $\varepsilon_6^{(E)}$, which correspond to the two-dimensional irreducible representation of the unitary subgroup C_{3V},

$$a_{ij} = \begin{bmatrix} a_{\xi\xi} & 0 & a_{\xi z} \\ 0 & a_{\eta\eta} & a_{\eta z} \\ (a_{\xi z})^* & (a_{\eta z})^* & 0 \end{bmatrix} , \tag{55}$$

where for the case of the acoustic magnons the components of (55) are determined by the expressions

$$a_{\xi z}^{(5,\mathrm{I})}=-a_{\xi z}^{(5,\mathrm{II})}=a_{\eta z}^{(5,\mathrm{I})}=a_{\eta z}^{(5,\mathrm{II})}=-\left[\frac{3S}{2}\right]^{1/2}\left\{i(\lambda_3-\lambda_4)\left[\frac{\tilde{A}_5}{\tilde{I}_5}\right]^{1/4}+\frac{3S}{2}P_{11}\left[\frac{\tilde{I}_5}{\tilde{A}_5}\right]^{1/4}\sin2\vartheta\right] ,$$

$$a_{\xi\xi}^{(5,\mathrm{I})}=a_{\xi\xi}^{(5,\mathrm{II})}=-a_{\eta\eta}^{(5,\mathrm{I})}=a_{\eta\eta}^{(5,\mathrm{II})}=-3S\left[\frac{3S}{2}\right]^{1/2}P_4\left[\frac{\tilde{A}_5}{\tilde{I}_5}\right]^{1/4}\cos\vartheta ,$$

and for the exchange magnons

$$a_{\xi z}^{(6,\mathrm{I})}=-a_{\xi z}^{(6,\mathrm{II})}=-a_{\eta z}^{(6,\mathrm{I})}=-a_{\eta z}^{(6,\mathrm{II})}=\left[\frac{3S}{2}\right]^{1/2}\left\{-i[(\lambda_3+\lambda_4)\sin\vartheta-\lambda_5\cos\vartheta]\left[\frac{\tilde{I}_6}{\tilde{i}_6}\right]^{1/4}+3SP_{11}\left[\frac{\tilde{i}_6}{\tilde{I}_6}\right]^{1/4}\cos\vartheta\right\} ,$$

$$a_{\xi\xi}^{(6,\mathrm{I})}=a_{\xi\xi}^{(6,\mathrm{II})}=a_{\eta\eta}^{(6,\mathrm{I})}=-a_{\eta\eta}^{(6,\mathrm{II})}=\left[\frac{3S}{2}\right]^{3/2}(P_4-2P_5)\left[\frac{\tilde{I}_6}{\tilde{i}_6}\right]^{1/4}\sin2\vartheta .$$

It follows from formulas (53) that in contrast to the phase T_2 the exchange mode corresponding to the one-dimensional representation is not silent any more. The tensor of scattering for this mode is determined by the quadratic magneto-optic constants of the exchange nature. However, all the components of the tensor contain the small factor $\sin\vartheta$. In distinction to the tensor of scattering from the acoustic mode corresponding to the one-dimensional representation, all the components of the tensor of scattering from the acoustic magnons corresponding to the two-dimensional representation contain the contribution of the exchange mechanism of the scattering. This contribution is exchange enhanced in the $a_{\xi z}$ and $a_{\eta z}$ components despite the presence of the small factor $\sin\vartheta$. Finally, the tensor of scattering from the exchange magnons corresponding to the two-dimensional representation does not tend to zero even in the exchange approximation in the same manner as for the T_2 magnetically ordered phase.

Let us turn to the consideration of the influence of the dipole-dipole interaction on the magnon spectrum and selection rules in the magnetically ordered phase T_3. Note that besides the magnon $\varepsilon_1^{(E)}$ all five other magnons are magneto-dipole active in the given phase. Using the relationships (45)–(47) and (52), one can write the expression for the Hamiltonian $\mathcal{H}_{d\text{-}d}$ in the ordinary Cartesian coordinate system for the case of the arbitrarily oriented vector

$$\mathbf{f}=(\cos\phi_{\mathrm{q}}\sin\theta_{\mathrm{q}},\sin\phi_{\mathrm{q}}\sin\theta_{\mathrm{q}},\cos\theta_{\mathrm{q}})$$

in the form

$$\mathcal{H}^{(2)}_{d\text{-}d}(\mathbf{q})=\{g_E[\cos\phi_{\mathbf{q}}(\xi^\dagger_{6,\mathrm{I}}+\xi_{6,\mathrm{I}})-i\sin\phi_{\mathbf{q}}(\xi^\dagger_{6,\mathrm{II}}-\xi_{6,\mathrm{II}})]\sin\theta_{\mathbf{q}}$$
$$+g_{A_5}[-\cos\phi_{\mathbf{q}}(\xi^\dagger_{5,\mathrm{II}}+\xi_{5,\mathrm{II}})+i\sin\phi_{\mathbf{q}}(\xi^\dagger_{5,\mathrm{I}}-\xi_{5,\mathrm{I}})]\sin\theta_{\mathbf{q}}+g_{A_4}(\xi^\dagger_4+\xi_4)\cos\theta_{\mathbf{q}}\}^2 \ . \tag{56}$$

In this expression

$$g_E=\mu_B\left[\frac{3\pi S}{v_0}\right]^{1/2}(g^{(1)}_{xx}\sin\vartheta-g^{(1)}_{xz}\cos\vartheta)\left[\frac{\tilde{I}_6}{\tilde{i}_6}\right]^{1/4},$$

$$g_{A_5}=\mu_B\left[\frac{3\pi S}{v_0}\right]^{1/2}g^{(1)}_{yy}\left[\frac{\tilde{A}_5}{\tilde{I}_5}\right]^{1/4},$$

$$g_{A_4}=\mu_B\left[\frac{6\pi S}{v_0}\right]^{1/2}(g^{(1)}_{zx}\sin\vartheta-g^{(1)}_{zz}\cos\vartheta)\left[\frac{\tilde{A}_4}{\tilde{I}_4}\right]^{1/4}.$$

The solution of the dispersion equation taking into account the contribution from $\mathcal{H}^{(2)}_{d\text{-}d}$ shows that at any orientation of the vector $\mathbf{q}$ there exist two transverse magnons, one exchange and one acoustic. The direction of the magnetic-moment oscillations corresponding to the acoustic magnon is always oriented in the XY plane and at the same time normal to the vector $\mathbf{q}$. These magnons have the energies $\Omega_{E\perp}=\varepsilon_{E_6}$ and $\Omega_{A\perp}=\varepsilon_{A_5}$. The energies of the remaining magnons are the solutions of the equation

$$(\Omega^2-\varepsilon^2_{E\parallel})(\Omega^2-\varepsilon^2_{A\parallel})(\Omega^2-\varepsilon^2_{A_4})$$
$$-4(\Omega^2-\varepsilon^2_{E\parallel})(\Omega^2-\varepsilon^2_{A\parallel})g^2_{A_4}\varepsilon_{A_4}\cos^2\theta_{\mathbf{q}}$$
$$-4(\Omega^2-\varepsilon^2_{E\parallel})(\Omega^2-\varepsilon^2_{A_4})g^2_{A_5}\varepsilon_{A_5}\sin^2\theta_{\mathbf{q}}$$
$$-4(\Omega^2-\varepsilon^2_{A\parallel})(\Omega^2-\varepsilon^2_{A_4})g^2_E\varepsilon_{E_6}\sin^2\theta_{\mathbf{q}}=0 \ . \tag{57}$$

Here

$$\varepsilon^2_{E\parallel}=\varepsilon^2_{E_6}+4g^2_E\varepsilon_{E_6}\sin^2\theta_{\mathbf{q}} \ ,$$

$$\varepsilon^2_{A\parallel}=\varepsilon^2_{A_5}+4g^2_{A_5}\varepsilon_{A_5}\sin^2\theta_{\mathbf{q}} \ .$$

It follows from this that the degeneration of the states of the pair of exchange and acoustic magnons remains only for the case $\mathbf{q}\parallel 0Z$ (at $\theta_{\mathbf{q}}=0$). The mixing of the states of the magnons corresponding to the two-dimensional and one-dimensional magnetodipole-active representations at $0<\theta_{\mathbf{q}}<\pi/2$ leads to the appearance of oblique polarizations lying in the $\mathbf{q}0Z$ plane of all three magnons. The pure longitudinal exchange and acoustic magnons appear at $q\perp 0Z$ (at $\theta_{\mathbf{q}}=\pi/2$) only.

The mixing of the states of these longitudinal magnons can be neglected in the case of the fulfillment of the condition $\varepsilon_{E_6}\gg\varepsilon_{A_5}$. In this case the longitudinal-transverse splitting is supposedly more significant for the exchange magnons

$$\Omega^2_{E\parallel}-\Omega^2_{E\perp}=8g^2_E\varepsilon_{E_6} \ . \tag{58}$$

The transition to the operators $\zeta^\dagger$ and ζ of the creation and annihilation of the longitudinal and transverse magnons is determined by the following relations:

$$\begin{bmatrix}\Psi_{6,\mathrm{I}}+\Psi_{6,\mathrm{III}}\\ \Psi_{5,\mathrm{I}}-\Psi_{5,\mathrm{III}}\\ \Psi_{6,\mathrm{V}}\end{bmatrix}=\exp(i\phi_{\mathbf{q}})\sqrt{6S}\,\hat{p}\begin{bmatrix}-\left[\frac{\tilde{I}_5}{\tilde{A}_5}\right]^{1/4}\left[\zeta^\dagger_{A\perp}+\zeta_{A\perp}+i\left[\frac{\Omega_{A\parallel}}{\varepsilon_{A_5}}\right]^{1/2}(\zeta^\dagger_{A\parallel}+\zeta_{A\parallel})\right]\\ \left[\frac{\tilde{A}_5}{\tilde{I}_5}\right]^{1/4}\left[\zeta^\dagger_{A\perp}-\zeta_{A\perp}+i\left[\frac{\varepsilon_{A_5}}{0_{A\parallel}}\right]^{1/2}(\zeta^\dagger_{A\parallel}-\zeta_{A\parallel})\right]\\ 0\end{bmatrix},$$

$$\begin{bmatrix}\Psi_{5,\mathrm{I}}+\Psi_{5,\mathrm{III}}\\ \Psi_{6,\mathrm{I}}-\Psi_{6,\mathrm{III}}\\ \Psi_{5,\mathrm{V}}\end{bmatrix}=\exp(i\phi_{\mathbf{q}})\sqrt{6S}\,\hat{p}\begin{bmatrix}\left[\frac{\tilde{I}_6}{\tilde{i}_6}\right]^{1/4}\left[i(\zeta^\dagger_{E\perp}+\zeta_{E\perp})+\left[\frac{\varepsilon_{E_6}}{\Omega_{E\parallel}}\right]^{1/2}(\zeta^\dagger_{E\parallel}+\zeta_{E\parallel})\right]\\ -\left[\frac{\tilde{i}_6}{\tilde{I}_6}\right]^{1/4}\left[i(\zeta^\dagger_{E\perp}-\zeta E\perp)+\left[\frac{\Omega_{E\parallel}}{\varepsilon_{E_6}}\right]^{1/2}(\zeta^\dagger_{E\parallel}-\zeta_{E\parallel})\right]\\ 0\end{bmatrix}, \tag{59}$$

in which the quantity $\Omega_\parallel$ is determined by (58).

Let us consider the form of the scattering tensors and the angle dependencies which appear in the geometry when the scattering plane coincides with the XY plane. The orientations of the vectors of incident and scattered light are shown in Fig. 4. In the case when the polarization of the incident light $\mathbf{e}^{(i)}\perp 0Z$ and polarization of the scattered light $\mathbf{e}^{(s)}\parallel 0Z$ the intensity of the scattering $I_{(\mathrm{sc})}$ is determined by the off-diagonal components a_{nd} of tensor (55) and depends only on the scattering angle ψ. For the scattering from the longitudinal magnons we have

$$I^{\nu\parallel}_{(\mathrm{sc})}\sim|a^{\nu\parallel}_{nd}|^2\sin^2(\psi/2)$$

and for the scattering from the transverse magnons

$$I^{\nu\perp}_{(\mathrm{sc})}\sim|a^{\nu\perp}_{nd}|^2\cos^2(\psi/2) \ .$$

Thus in the case of backward scattering at $\mathbf{q}\perp 0Z$ the intensity of the scattering from the transverse magnons turns to zero. The situation here is similar to the one that took place in the case of UO_2.

The picture is slightly more complicated if $\mathbf{e}^{(i)}\perp 0Z$ and $\mathbf{e}^{(s)}\perp 0Z$. The intensity of scattering is determined by the diagonal a_d components of the tensor (55) in this case. For the scattering from the transverse magnons we have

$$I_{(\mathrm{sc})}^{\nu\perp} \sim |a_d^{\nu\perp}|^2\cos^2 3(\phi+\psi/2)$$

and for the scattering from the longitudinal ones

$$I_{(\mathrm{sc})}^{\nu\parallel} \sim |a_d^{\nu\parallel}|^2\sin^2 3(\phi+\psi/2) \ .$$

The intensity of the scattering depends on the angle ϕ between the $0X$ axis and the wave vector of the incident light for these polarizations. However, this intensity does not depend on the choice of the orientation of the $0X$ axis within the plane normal to the $0Z$ axis.

The form of the obtained angular dependences does not depend on taking into account the mixing of the states of acoustic and exchange magnons. If one neglects the mixing one can obtain for the scattering from the transverse magnons

$$|a_{nd}^{E\perp}| = |a_{\xi z}^{(6,1)}|, \quad |a_{nd}^{A\perp}| = |a_{\xi z}^{(5,1)}| \ ,$$
$$|a_{d}^{E\perp}| = |a_{\xi\xi}^{(6,1)}|, \quad |a_{d}^{A\perp}| = |a_{\xi\xi}^{(5,1)}| \ .$$

The presence of the factors $(\varepsilon/\Omega)^{\pm 1/2}$ in formulas (59) slightly changes the form of the components of the tensors of scattering from the longitudinal magnons, for example,

$$a_{nd}^{E\parallel} = \left[\frac{3S}{2}\right]^{1/2} \left\{ -i\left[(\lambda_3+\lambda_4)\sin\vartheta - \lambda_5\cos\vartheta\right] \left[\frac{\tilde{I}_6}{\tilde{i}_6}\right]^{1/4} \left[\frac{\varepsilon_{E_6}}{\Omega_{E\parallel}}\right]^{1/2} + 3SP_{11}\left[\frac{\tilde{i}_6}{\tilde{I}_6}\right]^{1/4} \left[\frac{\Omega_{E\parallel}}{\varepsilon_{E_6}}\right]^{1/2} \cos\vartheta \right\}$$

and for the diagonal components

$$|a_d^{E\parallel}| = |a_{\xi\xi}^{(6,1}| \left[\frac{\varepsilon_{E_6}}{\Omega_{E\parallel}}\right]^{1/2} .$$

One can easily obtain analogous formulas for the components of the tensors of scattering from the acoustic longitudinal magnon with the help of (59). Note that all the peculiarities connected with the contribution of the exchange mechanism of scattering will remain as before.

The analysis of the influence of the dipole-dipole interaction on the behavior of the spectra of scattering in fact reduces to correctly taking into account the additional symmetry of the problem which is generated by the appearance of the magnon wave vector. This additional symmetry is connected with the orientation of the magnon wave vector in a crystal of the given symmetry. It is easy to see that if we do not take into account the dipole-dipole interaction under the condition when the twice-degenerate states of acoustic and exchange magnons are indistinguishable with respect to the energy all the angular dependencies will be absent for the above-considered geometry of scattering. That is why it is of interest to carry out the experiment to observe these angular dependencies. Their detection can testify to the considerably large value of the longitudinal-transverse splitting.

V. MAGNETIC STRUCTURE OF THE Nd_2CuO_4 TYPE

Consider the light scattering from magnons in the four-sublattice exchange-noncollinear antiferromagnet Nd_2CuO_4. The results published earlier[8] will be amplified by taking into account the dipole-dipole interaction and furthermore by the consideration of the peculiarities of the manifestation of the exchange mechanism of scattering in the presence of an external magnetic field.

In the stoichiometric composition Nd_2CuO_4 is a four-sublattice exchange-noncollinear antiferromagnet with the ordering temperature $T_N = 276$ K. Nd_2CuO_4 possesses a unique magnetic structure of the "plane cross" type in the magnetically ordered state.[13–15] The existence of this peculiar magnetic ordering in Nd_2CuO_4 may be considered as reliably revealed by measurements in an external magnetic field.[15–17] The exchange-noncollinear magnetic phase which exists in the temperature intervals[13–15] $1.5 < T < 30$ K and $75\ \mathrm{K} < T < T_N$ is shown in Fig. 5. We will restrict ourselves to the consideration of this magnetic phase only.

The structural phase transition

$$D_{4h}^{17}\ (I4/mmm) \Longrightarrow D_{4h}^{14}\ (P4_2/mnm)$$

taking place at $T = 300$ K precedes the magnetic ordering of the spins of the copper ions.[13,14,6] The results of Ref. 18 in which the light scattering from the electron transitions between the basic multiplet $^4I_{9/2}$ and the first excited multiplet $^4I_{11/2}$ of the Nd^{3+} ion has been studied can testify in favor of this phase transition. A doublet structure of spectra in the temperature interval $10 < T < 150$ K has been observed. The authors of Ref. 18 interpreted this structure of spectra as the so-called Davydov splitting phenomenon based on the fact that the symmetry group of Nd_2CuO_4 is D_{4h}^{17} $(I4/mmm)$ and the unit cell contains two neodymium ions occupying positions with the local symmetry C_{4V}. However, if one considers that the structural phase transition $D_{4h}^{17} \Longrightarrow D_{4h}^{14}$ did occur the shift of the copper ions has to be necessarily accompanied by displacements of the neodymium ions. In this case the

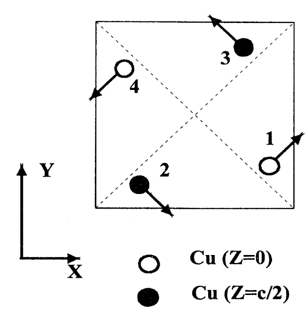

FIG. 5. Magnetic phase τ_2 of Nd_2CuO_4.

number of neodymium ions in the unit cell becomes equal to 8 and their positional symmetry lowers to C_s.[6] The last circumstance has evidently been manifested in the spectra of scattering observed in Ref. 18. Unlike the electron spectra of scattering, the phonon ones are less sensitive to the lowering of the local symmetry and the presence of the structural distortions because of the small absolute value of the last.

It is necessary, however, to note that the exchange-noncollinear magnetic structure shown in Fig. 5 can exist in both the presence and absence of structural distortions. In what follows we will assume that structural distortions leading to copper ion displacements are present and the symmetry of the paramagnetic phase is D_{4h}^{14}. The magnetic representation of the given group is realized by the components of the vectors which are linear combinations of the copper ion spins of the form (3). The classification of the components of these vectors from (3) in accordance with the irreducible representation of this group is given in Table III. Here and further we are using the same numbering of the copper ions (see Fig. 5) and the frequencies of magnetic resonance as used in Refs. 9 and 12.

Based on Table III one can represent the Hamiltonian of the magnetic subsystem of the copper ions in the form

$$\begin{aligned}\mathcal{H}=&J_0\mathbf{F}^2+J_1(\mathbf{L}_1^2+\mathbf{L}_2^2)+a_2(L_{1x}+L_{2y})^2+a_4(L_{1y}-L_{2x})^2+a_6(L_{1x}-L_{2y})^2+a_8(L_{1y}+L_{2x})^2+d(F_xL_{3y}+F_yL_{3x})+aL_{3z}^2\\&+\tfrac{1}{16}D[(\mathbf{L}_1^2-\mathbf{L}_2^2)^2+(\mathbf{F}_1^2-\mathbf{L}_3^2)^2-8(\mathbf{F}\cdot\mathbf{L}_1)^2-8(\mathbf{F}\cdot\mathbf{L}_2)^2]-H_x(g_{xx}F_x+g_{xy}L_{3y})-H_y(g_{yy}F_y+g_{yx}L_{3x})-g_{zz}H_zF_z\ ,\end{aligned}\tag{60}$$

where J, D, d, and a are the constants of bilinear exchange, four-spin exchange, Dzyaloshinskii-Moriya interaction, and anisotropy, respectively. The explicit form of these constants written in terms of the constants of intersublattice interactions is given in Ref. 9. The following equilibrium values of the linear combinations of the copper spins (3)

$$\bar{L}_{1x}=\bar{L}_{2y}=\sqrt{8}S\ ,$$

which are transformed in accordance with the τ_2 irreducible representation of the D_{4h}^{14} group correspond to the magnetic phase under consideration. The given magnetically ordered phase is stable under the conditions[6]

$$D>0,\quad a_2<0,a_4,a_6,a_8\ .$$

The transition to the local coordinate systems which are necessary for the calculations of the magnon energies is determined by the relations

$$\begin{aligned}&\begin{bmatrix}F_x\\L_{3y}\\L_{2z}\end{bmatrix}=\hat{p}\begin{bmatrix}L'_{2x}\\L'_{2y}\\L'_{1z}\end{bmatrix},\quad\begin{bmatrix}L_{3x}\\F_y\\L_{1z}\end{bmatrix}=\hat{p}\begin{bmatrix}L'_{1x}\\L'_{1y}\\L'_{2z}\end{bmatrix},\\&\begin{bmatrix}L_{2x}\\L_{1y}\\F_z\end{bmatrix}=\hat{p}\begin{bmatrix}F'_x\\F'_y\\L'_{3z}\end{bmatrix},\quad\begin{bmatrix}L_{1x}\\L_{2y}\\L_{3z}\end{bmatrix}=\hat{p}\begin{bmatrix}L'_{3x}\\L'_{3y}\\F'_z\end{bmatrix},\end{aligned}\tag{61}$$

where the matrix $\hat{p}$ has the form

$$\hat{p}=\begin{bmatrix}1/\sqrt{2}&0&1/\sqrt{2}\\1/\sqrt{2}&0&1/\sqrt{2}\\0&1&0\end{bmatrix}.\tag{62}$$

After the substitution of (61) and (10) into (60) it is easy to see that the part of the Hamiltonian $\mathcal{H}^{(2)}$ quadratic with respect to the operators of spin deviations takes the canonical form

$$\mathcal{H}^{(2)}=\tfrac{1}{2}\sum_L(q_LQ_L^2-p_LP_L^2)\ .\tag{63}$$

Here L runs from 0 to 3 which corresponds to F, L_1, L_2, and L_3. The quantities q_L and p_L are determined by the formulas

$$\begin{aligned}&p_0=8S(J_0-J_1),\quad p_1=p_2=-16Sa_2\ ,\\&p_3=-8S(J_1+2a_2+a)\ ,\\&q_0=16S(a_4-a_2)\ ,\\&q_1=q_2=8S(J_0-2J_1-4S^2D-d-4a_2)\ ,\\&q_3=8S[4S^2D+2(a_6-a_2)]\ .\end{aligned}\tag{64}$$

The group D_4 is the unitary subgroup for the magnetically ordered phase τ_2 in Nd_2CuO_4. The energies of the two acoustic magnons corresponding to the two-dimensional representation of this group are degenerate, $\varepsilon_{A_1}=\varepsilon_{A_2}=\sqrt{p_1q_1}$. The energy of the third acoustic magnon $\varepsilon_{A_3}=\sqrt{p_0q_0}$ is lower than that of the first two. This is connected with the fact that the expression for p_1 contains a strong intralayer anisotropy which prevents the copper spins from leaving the XY plane, and that the expression for q_0 contains only a weak interlayer anisotropy. This circumstance has been discussed in Refs. 9

TABLE III. Classification of the components of the vectors **L** in accordance with the irreducible representations of the group D_{4h}^{14}.

D_{4h}^{14}	
τ_1; A_{1g}	
τ_3; A_{2g}	F_z
τ_5; B_{1g}	L_{3z}
τ_7; B_{2g}	
τ_9; E_g	$\{F_x, \{L_{3y}$ F_y, L_{3x}
τ_2; A_{1u}	$L_{1x}+L_{2y}$
τ_4; A_{2u}	$L_{1y}-L_{2x}$
τ_6; B_{1u}	$L_{1x}-L_{2y}$
τ_8; B_{2u}	$L_{1y}+L_{2x}$
τ_{10}; E_u	$\{L_{2z}$ L_{1z}

and 12 in greater detail. The value of the energy of the exchange magnon $\varepsilon_E=\sqrt{p_3 q_3}$ is determined by the constant of the four-spin exchange D for which the relationship

$$J_1 \gg D \tag{65}$$

is satisfied. Any measurements of the value of the energy ε_E are not known to us so far.

The transition to the operators $\xi_\nu^\dagger$ and ξ_ν of the creation and annihilation of the magnons is determined by the relations

$$L_x'=\sqrt{2S}\, t_{L\nu}(\xi_\nu^\dagger+\xi_\nu),\quad L_y'=i\sqrt{2S}\, d_{L\nu}(\xi_\nu^\dagger-\xi_\nu)\ , \tag{66}$$

where

$$t_{L\nu}=\left[\frac{p_L}{q_L}\right]^{1/4},\quad d_{L\nu}=\left[\frac{q_L}{p_L}\right]^{1/4}.$$

Thus along with the exchange-enhanced coefficients d_{1A_1}, d_{2A_2}, and t_{0A_3} for the acoustic magnons the coefficient t_{3E} for the exchange magnon will also be exchange enhanced due to the relation (65).

Consider now the spin-dependent part of the dielectric permittivity $\Delta\varepsilon_{ij}$. We keep only those terms quadratic with respect to the operators $\mathbf{L}$ which have the exchange nature

$$\begin{aligned}
&\Delta\varepsilon_{xx}=\sigma_0\mathbf{F}^2+\sigma_1\mathbf{L}_1^2+\sigma_2\mathbf{L}_2^2\ ,\\
&\Delta\varepsilon_{yy}=\sigma_0\mathbf{F}^2+\sigma_2\mathbf{L}_1^2+\sigma_1\mathbf{L}_2^2\ ,\\
&\Delta\varepsilon_{zz}=\sigma_3\mathbf{F}^2+\sigma_4(\mathbf{L}_1^2+\mathbf{L}_2^2)\ ,\\
&\Delta\varepsilon_{xy}=\lambda_3 F_z+\sigma_5(\mathbf{L}_1\cdot\mathbf{L}_2)\ ,\\
&\Delta\varepsilon_{yz}=\lambda_1 F_x+\lambda_2 L_{3y},\quad \Delta\varepsilon_{yz}=-\lambda_1 F_y-\lambda_2 L_{3x}\ .
\end{aligned}\tag{67}$$

The orthogonality relations (7) for the vectors $\mathbf{L}_\beta$ ($\beta=0,1,2,3$) have been used while obtaining expressions (67). The coefficients σ may be expressed through the spin-dependent polarizabilities $\pi_{ijlm}^{\alpha\beta}$ of the pair of ions from α and β sublattices

$$\begin{aligned}
&\sigma_0=\tfrac{1}{6}\mathrm{Tr}(\pi_{xx11}^{12}+\pi_{xx11}^{13}),\quad \sigma_1=\tfrac{1}{6}\mathrm{Tr}(\pi_{xx11}^{12}-\pi_{xx11}^{14})\ ,\\
&\sigma_2=\tfrac{1}{6}\mathrm{Tr}(\pi_{xx11}^{13}-\pi_{xx11}^{14}),\quad \sigma_3=\tfrac{1}{3}\mathrm{Tr}(\pi_{zz11}^{12})\ ,\\
&\sigma_4=\tfrac{1}{6}\mathrm{Tr}(\pi_{zz11}^{12}-\pi_{zz11}^{14}),\quad \sigma_5=\tfrac{1}{3}\mathrm{Tr}(\pi_{xy11}^{14})\ .
\end{aligned}$$

If one substitutes the equilibrium values of the operators $\mathbf{L}$ into formulas (67) the change of the dielectric permittivity caused by magnetic ordering is obtained.

Substituting (61) and (66) into expressions (67) and using the definition (1) one can obtain the tensors of scattering from magnons for Nd_2CuO_4:

$$\begin{aligned}
&a^{A_1}=\begin{bmatrix}0&0&c\\0&0&0\\-c&0&0\end{bmatrix},\quad a^{A_2}=\begin{bmatrix}0&0&0\\0&0&c\\0&-c&0\end{bmatrix},\\
&a^{A_3}=\begin{bmatrix}0&b&0\\-b&0&0\\0&0&0\end{bmatrix},\quad a^{E}=\begin{bmatrix}d&0&0\\0&-d&0\\0&0&0\end{bmatrix}.
\end{aligned}\tag{68}$$

The b and c components of the tensors of scattering from the acoustic magnons contain the exchange-weakened constants of the linear magneto-optic effects,

$$c=(-\lambda_1+\lambda_2)\sqrt{S}\left[\frac{p_1}{q_1}\right]^{1/4},\quad b=i\lambda_3\sqrt{2S}\left[\frac{q_0}{p_0}\right]^{1/4}.$$

Similarly to the cases of UO_2 and $R$$MnO_3$ the scattering from the acoustic magnons in Nd_2CuO_4 is absent in the exchange approximation.

An absolutely different situation takes place in the case of scattering from the exchange magnons. The components of the scattering tensor contain the constants of the quadratic magneto-optic effects of the exchange nature:

$$d=(\sigma_2-\sigma_1)S\sqrt{8S}\left[\frac{-J_1-2a_2-a}{4S^2D+2(a_6-a_2)}\right]^{1/4}. \tag{69}$$

In contrast with all the cases considered above these magneto-optic constants are exchange enhanced. Thus a unique situation exists in Nd_2CuO_4, namely, the intensity of scattering from the exchange magnon mode will be several orders of magnitude larger than the intensity of scattering from the acoustic modes.

Consider the influence of the dipole-dipole interaction on the states and energies of the acoustic magnons. The exchange magnon is not magnetodipole active in this crystal. The tensor of the g factors for the first ion has the off-diagonal components

$$g_{ij}=\begin{bmatrix}g_{xx}&g_{xy}&0\\g_{xy}&g_{xx}&0\\0&0&g_{zz}\end{bmatrix}. \tag{70}$$

The equalities $g_{xx}=g_{yy}$ and $g_{xy}=g_{yx}$ are explicitly taken into account while writing the expression (70).

Using (70) and Table III, one can write the Hamiltonian $\mathcal{H}_{d\text{-}d}$ as

$$\begin{aligned}
\mathcal{H}_{d\text{-}d}=\frac{2\pi}{v_0}\mu_B^2\{&f_x(g_{xx}F_x+g_{xy}L_{3y})\\
&+f_y(g_{xx}F_y+g_{xy}L_{3x})+f_z g_{zz}F_z\}^2\ .
\end{aligned}\tag{71}$$

Here $\mathbf{f}=\mathbf{q}\cdot q^{-1}$. The part of $\mathcal{H}_{d\text{-}d}$ obtained after the transition to the operators $\xi_\nu^\dagger$ and ξ_ν of creation and annihilation of the magnons has the form

$$\begin{aligned}
\mathcal{H}_{d\text{-}d}^{(2)}=\{&-g_1 f_x(\xi_{A_2}^\dagger+\xi_{A_2})+g_1 f_y(\xi_{A_1}^\dagger+\xi_{A_1})\\
&+ig_3 f_z(\xi_{A_3}^\dagger-\xi_{A_3})\}^2\ ,
\end{aligned}\tag{72}$$

where

$$g_1=\left[\frac{8\pi S}{v_0}\right]^{1/2}\mu_B(g_{xx}-g_{xy})\left[\frac{p_1}{q_1}\right]^{1/4},$$

$$g_3=\left[\frac{16\pi S}{v_0}\right]^{1/2}\mu_B g_{zz}\left[\frac{q_0}{p_0}\right]^{1/4}.$$

The diagonalization of the Hamiltonian $\mathcal{H}+\mathcal{H}_{d\text{-}d}^{(2)}$ leads to the same situation which was realized for the T_3 magnetic phase of $R\mathrm{MnO}_3$. There always exists the transverse magnon $\Omega_{A1}=\varepsilon_{A_1}$, which corresponds to the oscillations of the magnetic moment lying in the XY plane, for any orientation of the wave vector $\mathbf{q}$. The polarizations of the other two magnons are oblique and lying in the $0Z\mathbf{q}$ plane. The energies of these magnons are determined by the equation

$$(\Omega^2-\varepsilon_{A_1}^2)(\Omega^2-\varepsilon_{A_2}^2)-(\Omega^2-\varepsilon_{A_1}^2)g_3^2\varepsilon_{A_3}\cos^2\theta_{\mathbf{q}} -(\Omega^2-\varepsilon_{A_3}^2)g_1^2\varepsilon_{A_1}\sin^2\theta_{\mathbf{q}}=0 .$$

In the case $\mathbf{q}\perp 0Z$ the polarization of one of the magnons is pure longitudinal and the polarization of the other one is pure transverse.

The coefficients of the unitary transformation from the states of the ordinary magnons to the states of the longitudinal-transverse magnons have the following form.

(a) For the transverse magnons

$$t_{A_1,\mu_\perp}=-\cos\varphi_{\mathbf{q}},\quad t_{A_2,\mu_\perp}=-\sin\varphi_{\mathbf{q}},\quad d_{A_3,\mu_\perp}=0 .$$

(b) For the other two magnons with the oblique polarization

$$t_{A_1,\mu}=\varepsilon_{A_1}g_1\sin\theta_{\mathbf{q}}\sin\varphi_{\mathbf{q}}(\Omega_\mu^2-\varepsilon_{A_3}^2)N_\mu ,$$

$$t_{A_2,\mu}=-\varepsilon_{A_1}g_1\sin\theta_{\mathbf{q}}\cos\varphi_{\mathbf{q}}(\Omega_\mu^2-\varepsilon_{A_3}^2)N_\mu ,$$

$$d_{A_3,\mu}=i\varepsilon_{A_3}g_3\cos\theta_{\mathbf{q}}(\Omega_\mu^2-\varepsilon_{A_1}^2)N_\mu .$$

In these formulas

$$N_\mu=\{\Omega_\mu[\varepsilon_{A_1}g_1^2(\Omega_\mu^2-\varepsilon_{A_3}^2)\sin^2\theta_{\mathbf{q}} +\varepsilon_{A_3}g_3^2(\Omega_\mu^2-\varepsilon_{A_1}^2)\cos^2\theta_{\mathbf{q}}]\}^{-1/2} .$$

Thus the form of the tensors of scattering becomes dependent on the scattering geometry if one takes into account the dipole-dipole interaction. This dependence is determined by the relation

$$a_{ij}^{\mu}=a_{ij}^{A_1}t_{A_1,\mu}+a_{ij}^{A_2}t_{A_2,\mu}+a_{ij}^{A_3}d_{A_3,\mu} . \qquad (73)$$

The analysis of this relation shows that at $\mathbf{q}\perp 0Z$ and $\mathbf{q}\|0Z$ in the geometry of backward scattering the only possible scattering is from the longitudinal magnons. It is similar to the foregoing cases of UO_2 and $R\mathrm{MnO}_3$.

Let us briefly discuss the influence of the external magnetic field on the behavior of the intensity of scattering and the changes of the form of the scattering tensor in $\mathrm{Nd_2CuO_4}$. We will be primarily interested in the appearance of the contributions of the exchange magneto-optic constants σ_0, σ_3, σ_4, and σ_5 which did not previously enter the tensor of scattering. However, we will not consider the region of the fields in which spin-reorientation phase transitions[9,11] take place. The behavior of the magnon modes of the copper magnetic subsystem in $\mathrm{Nd_2CuO_4}$ in an external magnetic field oriented along the highly symmetric directions has been considered in Ref. 9.

First of all it is necessary to note that all the newly appeared contributions and new components of the scattering tensors contain the induced magnetic moment $m\approx 4Sg\mu_B H/J_1$ as a factor in the region of magnetic fields of interest. The quantity m is small because the value J_1 contains the large interlayer exchange K_{14}. It is possible to show that in the case of a magnetic field oriented along the Z axis, $\mathbf{H}\|0Z$, all the magneto-optical constants of the exchange nature give the contribution to the components of the scattering tensor; however, besides the small factor m all these contributions contain the exchange-weakened t and d coefficients. Thus one can neglect the changes of the scattering tensors in this case.

For the magnetic field $\mathbf{H}\|0X$ mixing of the states of A_1, A_3 and A_2E magnons takes place. The tensor of the scattering from the A_2 and E modes has the form

$$a_{ij}^{A_2,E}=\begin{vmatrix} d_1 & 0 & 0 \\ 0 & d_2 & c_1 \\ 0 & c_2 & d_3 \end{vmatrix} .$$

The exchange-enhanced magneto-optic constant σ_0 gives a contribution to the components d_1 and d_2. The component d_3 contains the exchange-enhanced constant σ_3. However, these contributions contain the factor m^2. The tensor of the scattering from the A_1, A_3 modes contains the exchange-weakened contribution of the magneto-optic constant σ_5. An analogous situation takes place for the case $\mathbf{H}\|[110]$. Thus the role of the magneto-optic constants of the exchange nature, σ_0, σ_3, σ_4, and σ_5, is inessential in the case of light scattering in the presence of an external magnetic field.

The behavior of the scattering tensors in the region of the fields corresponding to the spin-reorientation phase transitions has to be considered separately.

VI. SUMMARY

The study of the Raman light scattering from magnons in many-sublattice exchange-noncollinear magnets carried out here has shown that these objects provide favorable situations for the detection and investigation of the exchange magnon modes by means of light scattering spectroscopy. It was shown that, in contrast with exchange-collinear magnets, the intensity of the scattering from some exchange magnons is determined by the magneto-optic constants of the exchange nature, as in hexagonal perovskites, and does not contain any small factors. It has also been demonstrated that, for the "plane cross" exchange-noncollinear magnetic ordering which exists in $\mathrm{Nd_2CuO_4}$, the components of the scattering tensor are proportional to the constants of the quadratic magneto-optic effects of the exchange nature, and what is more these constants are exchange enhanced. Due to this circumstance the intensity of the Raman light scattering from the exchange modes has to be several orders of magnitude higher than the intensity of the scattering from the acoustic modes.

Our approach allowed us to carry out an analysis of the experiments of Ref. 27. As was shown the Raman light scattering from the exchange magnon in UO_2 has actually been observed in Ref. 27. In exact correspondence with our theoretical results the observed intensity of scattering from the exchange magnon was higher than the intensity of scattering from the acoustic ones. As was shown in Sec. III the tensor of scattering from the exchange magnon in UO_2 is determined by the magneto-optic constant of the exchange nature and does not contain any small factors.

Let us recall that the intensity of the well-studied two-magnon light scattering is precisely determined by the quadratic magneto-optic constants of the exchange nature.[12,25] It is quite possible that the exchange magnons have already been observed in experiments on light scattering in exchange-noncollinear magnets but not, however, been identified, as in the case of UO_2.

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