Exchange mechanism of one-magnon light scattering

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The feasibility of one-magnon light scattering in ferro- and antiferromagnets is investigated within the framework of the exchange mechanism. A microscopic theory of this scattering is developed, its probability is calculated, and it is shown that the latter can differ from zero only if the exchange interactions in the ground state are not equal to those in the virtual excited states contributing to the scattering tensor. In this case there is no spin collinearity in the ground and excited states of an antiferromagnet placed in an external magnetic field that causes canting of the magnetic sublattices, or in a ferromagnet whose magnetization is not collinear with the external field. Dipole transitions can take place in this case between sublevels of the ground and excited multiplets with different spin projections, and as a consequence exchange interaction without rotation of the polarization plane becomes allowed. The intensity of this scattering has a nonmonotonic dependence on the applied field. The dielectric tensor of an antiferromagnet with anisotropy of the "easy plane" type is obtained, and the magnetic contributions to the refractive index of this antiferromagnet are determined.

Recent studies of Brillouin scattering of light by spin waves of antiferromagnetic (AFM) EuTe (Ref. 1, see also Ref. 2) have revealed for the first time one-magnon Raman scattering (RS) of light, due to exchange interaction. The principal distinctive feature of this RS is the equality of the polarizations of the incident and scattered photons, whereas the other known[3] microscopic mechanisms (magnetodipole[4] and spin-orbit[5]) of one-magnon RS alter the polarization of the scattered electromagnetic field. Constancy of the polarization of light inelastically scattered by AFM spin waves was heretofore observed only in second-order processes, when the scattering resulted either in creation (annihilation) of a pair of magnons, in a magnetically ordered medium, or else annihilation of one magnon with simultaneous creation of another,[6–9] so that the spin was preserved in the initial and final states. This led in fact to the conclusion that an interaction that does not lift the spin forbiddenness cannot serve as a source of first-order scattering or, in other words, of one-magnon RS.[6,8] Nonetheless, a more detailed examination of the role of exchange interaction shows that in certain situations it can also lead to one-magnon RS which, however, in contrast to the RS due to relativistic mechanisms, does not affect the polarization of the light wave. What is involved here is not only the ordinary exchange interaction that governs the static and dynamic properties of a magnet, but also exchange interaction between orbitally excited ions and non-excited ones,[1)] since it is precisely the virtual electrodipole (i.e., spin-allowed) transitions to orbitally excited states which determine the dielectric tensor and the magnitude of the scattering tensor in terms of which the RS cross section is expressed.[13] Since the exchange parameters for unexcited ions and for an ion pair one of which is excited (or else the parameters for the d–d, f–f and respectively s–d and d–f exchanges) are different, the spin configuration of the ground state of the AFM and a state with excited ions can likewise be unequal. (The first to suggest that the spin moments of the ion in its ground and excited states are not collinear were apparently Eremenko and co-workers[14] in an interpretation of the details of the MnF_2 exciton spectrum. They have also considered, with account taken of this circumstance, certain features of the light absorption by $NaMnCl_3$ at the frequencies of the one- and two-particle transitions in a magnetic field.[15]) The spin configurations are different, as will be shown below, only in an external field that disturbs the initial collinear spin configuration. The spin quantization axes in the ground and excited states are then different and the electrodipole transitions can couple states with unequal spin projections. In other words, inequality of the exchange-interaction parameters in the ground and virtual excited states, which play in the lifting of the spin forbiddenness a role similar to that of the spin-orbit interaction, determine the feasibility, in principle, of realizing an exchange RS mechanism with creation of one magnon, but manifest themselves only in the presence of an external field that causes the shear of the AFM sublattices.

We carry out below, in the exchange approximation (i.e., assuming absence of relativistic interactions) a microsocopic calculation, based on the theory of small-radius excitons in magnetic dielectrics, of the inelastic-light-scattering tensor and of the dielectric tensor of AFM. The actual calculations are carried out for an AFM model with anisotropy of the "easy plane" (EP) type (such as, in particular, EuTe) in an external magnetic field perpendicular to the symmetry axis.

SPIN CONFIGURATIONS

The scattering tensor and other optical characteristics are easy to calculate if the eigenstates of the ions in the crystal, exchange, and external fields are known. To simplify the exposition that follows we confine ourselves to two (ground and fth excited) ion terms having different parity and specified values of the spin S; the degeneracy of the latter is determined only by the multiplicity $2S + 1$. The latter does not make the analysis less general, since we are interested only in the role of the exchange interaction and neglect, as already mentioned, the spin-orbit interaction (generalization of the results of the case of a fine LSJ structure of the electronic terms of the ion, as well as allowance for the spin-orbit interaction, entails no difficulty in principle and can be carried out separately).

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Let, according to the foregoing, an AFM with exchange anisotropy of the EP type be placed in an external magnetic field $\mathbf{H}\perp Z$. The Hamiltonian that describes the ground state and the spin-wave spectrum of the AFM can then be written in the form

$$\mathcal{H}_S=\frac{1}{2}\sum_{n\alpha,\rho}(I\mathbf{S}_{n\alpha}\mathbf{S}_{n\alpha+\rho}-\Delta I S_{n\alpha}{}^{z}S^{z}_{n\alpha+\rho})-\mu_B gH\sum_{n\alpha}S_{n\alpha}{}^{x}, \quad (1)$$

where $I>\Delta I>0$ are the parameters of the exchange and of its anisotropy, the sign of which corresponds to the type of the EP, $\mathbf{n}$ is the cell vector, $\alpha(=1,2)$ is the number of the magnetic sublattice, $\mathbf{S}_{n\alpha}$ is the $n\alpha$ ion spin operator acting in the function space of the ground multiplet, g is the Lande factor, μ_B is the Bohr magneton, and ρ joins the nearest neighbors.

The total Hamiltonian of the AFM contains besides the operator (1) also the operator

$$\mathcal{H}^{(f)}=\sum_{n\alpha}h_{n\alpha}^{(f)}+\mathcal{H}_S^{(f)}+V_{res}^{(f)} \quad (2)$$

of the excited states. The first term of the latter includes the one-site operators in the crystal field, the second

$$\mathcal{H}_S^{(f)}=\varkappa|I_f|\sum_{n\alpha,\rho}\mathbf{S}_{n\alpha}^{(f)}\mathbf{S}_{n\alpha+\rho}-\mu_B g_f H\sum_{n\alpha}S_{n\alpha}^{(f)x} \quad (3)$$

describes the isotropic exchange interaction between the excited ion and the unexcited ions that surround it, and the operator $V_{res}^{(f)}$, the form of which will be given below, corresponds to the resonant (dipole-dipole) interaction that leads to collectivization of the electronic states in the crystal, i.e., to excitons.[9-12] The notation used in (2) is the following: I_f is the parameter of the exchange between the excited and unexcited ions, $\mathbf{S}_{n\alpha}^{(1)}$ is the spin operator and acts only on the functions of the excited multiplet, g_f is its Lande factor, $\varkappa\equiv\operatorname{sgn}I_f$ reflects the fact that the sign of I_f can be arbitrary.

To calculate the spin configurations of the ground and excited states, we change to the proper (local) coordinate frames. In the former case we have then

$$S_{n\alpha}{}^{x}=S_{n\alpha}{}^{\xi}\sin\theta+S_{n\alpha}{}^{\zeta}\cos\theta, \quad (4)$$

$$S_{n\alpha}{}^{y}=-(-1)^{\alpha}(S_{n\alpha}{}^{\xi}\cos\theta-S_{n\alpha}{}^{\zeta}\sin\theta), \quad S_{n\alpha}{}^{z}=-(-1)^{\alpha}S_{n\alpha}{}^{\eta},$$

and in the latter it is necessary to make the change $(-1)^{\alpha}\to\varkappa(-1)^{\alpha}$. The quantization axes will hereafter to be taken to be $O\zeta$ and $O\zeta_f$, and the angles between them and $\mathbf{H}$ are specified as θ and θ_f. The ground-state configuration for an AFM of the EP type is well known; it can be obtained here by substituting (4) in the operator (1), separating in the latter the one-ion part $\mathcal{H}_{n\alpha}$, and equating to zero the coefficient of the term linear in $S_{n\alpha}^{\xi}$. The dependence of the angle θ on $\mathbf{H}\perp Z$ in the region of the canted phase is then determined by the usual relation $\theta=\arccos(\mu_B gH/2\varepsilon_S)$ ($\varepsilon_S=Isz$, z is the number of nearest neighbors, and s is the average sublattice magnetization per site). The proper basis of the operator $\mathcal{H}_{n\alpha}=-\varepsilon_S S_{n\alpha}^{\zeta}$ is specified in this case by the set $|M;\zeta\rangle_{n\alpha}$, $|M|<S$, while the energies $M\varepsilon_S$ of the states of the ground multiplet in the exchange and external fields are independent of $\mathbf{H}$. Further diagonalization of the operator

$$H_S=\sum_{n\alpha}\mathcal{H}_{n\alpha}+V_S,$$

$$V_S=-\frac{1}{2}\sum_{n\alpha,\rho}[I\cos 2\theta S_{n\alpha}{}^{\xi}S_{n\alpha+\rho}^{\xi}+(I-\Delta I)S_{n\alpha}{}^{\eta}S_{n\alpha+\rho}^{\eta}], \quad (5)$$

is carried out in standard fashion.[3,16] In particular, the energies of the long-wave magnons for a cubic AFM take, as in Ref. 3, the form

$$\Omega_1{}^2(\mathbf{k})=\gamma^2H^2+\delta^2k, \quad \Omega_2{}^2(\mathbf{k})=2\gamma H_E H_A+\delta^2k^2,$$

where

$$\gamma=\mu_B g, \quad g\mu_B H_E=\varepsilon_S, \quad g\mu_B H_A=\Delta Isz, \quad 2\delta=a\varepsilon_S,$$

and $\mathbf{a}$ is the lattice vector.

A configuration with an excited atom corresponds to a free state of the operator (3), and since it is identical with the Hamiltonian of a collinear substitutional impurity in a magnetically ordered matrix,[17] the equilibrium value of the angle θ_f is also obtained similarly. Separating from (3) the one-ion Hamiltonian $h_{n\alpha}^{(f)}$, it is easy to obtain from the condition of its diagonalization in the proper axis system, the equation

$$\varepsilon_S{}^{(f)}\sin(\theta+\varkappa\theta_f)-\mu_B g_f H\sin\theta_f=0, \quad \varepsilon_S{}^{(f)}=|I_f|sz, \quad (6)$$

under the assumption that the rotation angles of the AFM spins are independent on their distances to the excited site. Strictly speaking, in a canted AFM the angles of the spins located near the excited ions differ from θ, but in magnetic dielectrics this difference is $\sim 1/z$ (Ref. 18) and we shall neglect it. Note that this approximation is not valid if $|I_f|\gg I$, when the excited ion and its nearest environment are strongly coupled. In this case the role of the "impurity" is assumed by an entire cluster, and θ_f can be regarded as the cluster-spin rotation angle. It is possible that this situation is closer to magnetic semiconductors, where the d–f exchange exceeds as a rule the exchange of f electrons with one another.[10]

From (6) we get

$$\cos\theta_f=\left(1-\frac{1}{2}\varkappa\frac{g}{g_f}\frac{\varepsilon_S^{(f)}}{\varepsilon_S}\right)\frac{\mu_B g_f H}{\varepsilon_S^{(f)}(\mathbf{H})}, \quad (7)$$

where

$$\varepsilon_S^{(f)}(\mathbf{H})=\left[(\varepsilon_S^{(f)})^2+\left(1-\varkappa\frac{g}{g_f}\frac{\varepsilon_S^{(f)}}{\varepsilon_S}\right)(\mu_B g_f H)^2\right]^{1/2} \quad (8)$$

determines the energies $M_f\varepsilon_S^{(f)}(\mathbf{H})$ of the spin sublevels of the excited multiplet in an external field. Comparing (7) and (8) we see that the influence of the exchange interaction of the surroundings on the behavior of the spin of the excited state cannot be reduced to a renormalization of g. In addition, it follows from (6) that θ depends on $\varkappa$. At $\varkappa=-1$ the coefficient in the expression for $\cos\theta_f$ is positive, $0\leqslant\theta_f\leqslant\pi/2$, and the angle between the quantization axes in the ground and excited states is $\beta_{\mathbf{H}}=\theta+\theta_f$. At $\varkappa=1$ the angle $\beta_{\mathbf{H}}=\theta-\theta_f$, and situations $0\leqslant\theta_f<\theta\leqslant\pi/2$ if $g\varepsilon_S^{(f)}<g_f\varepsilon_S$, $0\leqslant\theta<\theta_f\leqslant\pi/2$ if $g_f\varepsilon_S\leqslant g\varepsilon_S^{(f)}\leqslant 2g_f\varepsilon_S$, and $0\leqslant\theta\leqslant\pi/2\leqslant\theta_f\leqslant\pi$, if $g\varepsilon_S^{(f)}>2g_f\varepsilon_S$ are possible. In the latter case the parameter I_f turns out to be so large that the quantization axis of the excited-multiplet spin turns away from the field, and the value of $\varepsilon_S^{(f)}(\mathbf{H})$ decreases. We find thus, in contrast to Ref.

15, that in all cases $\theta_f \neq 0$ only at $\mathbf{H} \neq 0$, and $\theta = 0$ or π at $H \geqslant 2H_E$, when $\theta = 0$. In other words, a nontrivial spin structure of a one-ion excited (or impurity[17]) state can occur only in an uncorrelated AFM.

FIRST-ORDER SCATTERING TENSOR

The solutions of the Schrödinger equations for the one-ion Hamiltonians $\mathscr{H}_{n\alpha}$ of the ground term and $\mathscr{H}_{n\alpha}^{(f)} = h_{n\alpha}^{(f)} + h_{n\alpha}^{(S)}$ of the excited term are functions of $|M;\zeta\rangle_{n\alpha}$ and $|M_f;\zeta_f\rangle_{n\alpha}$, which constitute a complete set and pertain to their "own" coordinate systems. This means that the correct representation is

$$\mathscr{H}_{n\alpha} = -\varepsilon_S \sum_{n\alpha} \sum_{M} MP_{n\alpha}(M),$$

$$\mathscr{H}_{n\alpha}^{(f)} = \sum_{n\alpha} \sum_{M_f} [\mathscr{E} - M_f \varepsilon_S^{(f)}(H)] P_{n\alpha}^{(f)}(M_f), \tag{9}$$

where

$$P_{n\alpha}(M) = |M;\zeta\rangle_{n\alpha}\langle\zeta;M|, \quad P_{n\alpha}^{(f)}(M_f) = |M_f;\zeta_f\rangle_{n\alpha}\langle\zeta_f;M_f| \tag{10}$$

are the projection operators and $\mathscr{E}$ is the distance between the centers of gravity of the multiplets or the energy of the electron excitation of the ion in the paramagnetic phase. The same representation is convenient for the construction of the transition operators (Hubbard operators) that relate the sublevels of the two multiplets, viz., the creation operators

$$B_{n\alpha}^{+}(M_f;M) = |M_f;\zeta_f\rangle_{n\alpha}\langle\zeta;M| \tag{11}$$

and their Hermitian adjoints, the annihilation operators $B_{n\alpha}(M;M_f)$, the action and commutation properties of which are clear from the definition.

Using (11), we write down the interaction $W(t)$ of an AFM with the electric field $\mathbf{E}$ of an electromagnetic wave of frequency ω and wave vector $\mathbf{Q}$. At optical frequencies it takes the form

$$W(t) = -\frac{\mathscr{E}}{2\omega} \sum_{n\alpha} \sum_{M,M_f} (\mathbf{E}\mathbf{d}_\alpha)\langle\zeta_f;M_f|M;\zeta\rangle [B_{n\alpha}^{+}(M_f;M) - B_{n\alpha}(M;M_f)] e^{i(\mathbf{Q}n\alpha - \omega t)} + \text{c.c.}, \tag{12}$$

where $\mathbf{d}_\alpha$ is the dipole moment of the transition of an αth sublattice ion from the ground state to an excited ones ($\mathbf{d}_\alpha = \mathbf{d}$ in crystals in which the multisublattice structure is due to the magnetic ordering). In contrast to the collinear case,[11] here $M_f \neq M$ and the matrix elements $\langle\zeta_f;M_f|M;\zeta\rangle$ (the "overlap integrals" of the spin functions) are Wigner D-functions or the coefficients of the unitary transformation

$$|M;\zeta\rangle_{n\alpha} = \sum_{M_f} d_{M_f M}^{S}(\beta_H)\,|M_f;\zeta_f\rangle,$$

$$\sum_{M_f} d_{M_f M}^{S}(\beta_H)\, d_{M_f M'}^{S}(\beta_H) = \delta_{MM'} \tag{13}$$

(in the general case the D functions depend on the three Euler angles that specify the rotation of the $\xi\eta\zeta$ frame relative to $\xi_f\eta_f\zeta_f$ (Ref. 20).

The sought-for tensor $c_{if}^{(S)}$ of RS with spin-wave excitation can be easily obtained, according to Ref. 13, in second-order perturbation theory in the operator (12), and is determined in the present case by the matrix elements

$$w_\alpha(M' \leftarrow M) = 2\pi(\omega_{sc}\omega_{in})^{1/2} \sum_{M_f} d_{M_f M'}^{S}(\beta_H)\, d_{M_f M}^{S}(\beta_H) \times \left[\frac{(\mathbf{e}_{sc}\mathbf{d}_\alpha)(\mathbf{e}_{in}\mathbf{d}_\alpha)}{\omega_{in} - \mathscr{E}_{M_f M}} - \frac{(\mathbf{e}_{in}\mathbf{d}_\alpha)(\mathbf{e}_{sc}\mathbf{d}_\alpha)}{\omega_{sc} - \mathscr{E}_{M_f M}}\right], \tag{14}$$

where $\mathbf{e}_{in}$, $\mathbf{e}_{sc}$ and ω_{in}, ω_{sc} are the polarization unit vectors and the frequencies of the incident and scattered photons, and $\mathscr{E}_{M_f M} = \mathscr{E} - M_f \varepsilon_S^{(f)}(H) + M\varepsilon_S$. It follows from (14) that if the energy of the intermediate states is independent of M_f then, as can be seen from (13), $w_\alpha(M' \leftarrow M) \sim \delta_{MM'}$ and the investigated RS is impossible. However, assuming this dependence to exist, we get

$$w_\alpha(M' \leftarrow M) = -2\pi(\omega_{sc}\omega_{in})^{1/2} e_{sc}^{\,i} c_{ij}^{(S)} e_{in}^{\,j} \sum_{M_f} M_f d_{M_f M'}^{S}(\beta_H)\, d_{M_f M}^{S}(\beta_H),$$

$$c_{ij}^{(S)} = d_\alpha^{\,i} d_\alpha^{\,j} \varepsilon_S^{(f)}(\mathbf{H})[(\omega_{in} - \mathscr{E})^{-2} + (\omega_{sc} + \mathscr{E})^{-2}]. \tag{15}$$

The expression for w_α $(M' \leftarrow M)$ is similar to the corresponding matrix element in the spin-orbit mechanism,[3,6,8,9] but here it depends significantly on $\mathbf{H}$, and the polarizations of the scattered and incident waves may become equal (e.g., for cubic and uniaxial crystals we have $c_{ij}^{(S)} \sim \delta_{ij}$). The probability of first-order RS by spin waves is directly connected with the values of w_α $(M-1 \leftarrow M)$ (and also $w_\alpha(M+1 \leftarrow M)$ if the spin wave is annihilated). It is easy to find from the sum (15) by using the explicit form of the D-functions[20] and the dependences of the angles θ and θ_f on the external field, that the sought matrix elements (disregarding the numerical factor ~ 1 that depends on the value of S) are proportional to

$$\sin\beta_H = \frac{\varepsilon_S^{(f)}}{\varepsilon_S^{(f)}(\mathbf{H})}\left(\frac{g_f}{g}\frac{\varepsilon_S}{\varepsilon_S^{(f)}} - \varkappa\right)\sin 2\theta, \tag{16}$$

i.e., "exchange" RS is possible only in the noncollinear phase ($\theta \neq 0, \pi/2$). Note that noncollinearity can be due not to the applied field alone but, for example, to the Dzyaloshinskii interaction; then, in principle, the RS that conserves the electromagnetic-wave polarization can take place also in the absence of $\mathbf{H}$. A similar conclusion concerning the role of the noncollinearity follows also from a symmetry analysis of the components of the spin-dependent part of the dielectric tensor of a many-sublattice noncollinear orthoferrite.[21] Exchange RS is possible also in a ferromagnet if its magnetization and external field are not collinear.

Even the one-ion approximation used above demonstrates by itself a number of distinctive features of the exchange RS mechanism, such as its effectiveness for strongly canted spin structures and the diagonality of the scattering tensor. In this approximation, the dependence of the RS probability is determined in fact by the value of $\sin^2\beta_H$ (or $\sin^2 2\theta$), which is proportional to H^2 in weak ($H \ll H_E$) fields and the decreases to zero as the spin-flip field is approached (such a behavior accords qualitatively with the experiments[1]). Whereas, however, the exciton nature of the orbital states has little effect on the RS probability, this is not so for real spin excitations, viz., spin waves whose spectrum is significantly softened if the region of small wave vectors, which

is important for optical measurements. Furthermore, since the energies of the magnons depend in turn on the field, it becomes necessary to take into account the spatial dispersion of the spin excitations. At low temperatures, this can be done by introducing, following Refs. 6 and 8 (see also Ref. 9) the effective spin Hamiltonian

$$\mathscr{H}_{sc}^{\text{Stokes}}=(2S)^{-1/2}\mathbf{E}^2\sum_{n\alpha}w_\alpha S_{n\alpha}$$

of the Stokes RS, since the matrix element $w_\alpha\equiv w_\alpha(S-1\leftarrow S)$ relates the lowest and first-excited states of the ground multiplet in the proper coordinate frame (the matrix element and the corresponding operator for the anti-Stokes process can be expressed similarly). Transforming to the spin-wave representation[16] and putting $w_\alpha\equiv w$, we easily find that

$$\mathscr{H}_{sc}^{\text{Stokes}}=(2S)^{-1/2}w\mathbf{E}^2\sum_{\mathbf{k}}[u(\mathbf{k})+v(\mathbf{k})]\delta(\mathbf{Q}_{in}-\mathbf{Q}_{sc}+\mathbf{k})\beta^+(\mathbf{k})$$

$$\approx(2S)^{-1/2}w\mathbf{E}^2[u(0)+v(0)]\beta^+(0),$$

where $\mathbf{Q}_{in}$ and $\mathbf{Q}_{sc}$ are the wave vectors of the light before and after the scattering, $u(\mathbf{k})$ and $v(\mathbf{k})$ are Bogolyubov-Tyablikov transformation coefficients for the magnons of the acoustic mode, and $\beta^+(\mathbf{k})$ are the creation operators corresponding to them (at $\mathbf{H}\perp Z$ and $w_\alpha\equiv w$ the light is scattered only by acoustic magnons). For an AFM with EP anisotropy we have $u(0)+v(0)\approx(2\varepsilon|\Omega_1(0)|)^{1/2}=\cos^{-1/2}\theta$ (it is assumed here that the external field exceeds substantially the gap $(2H_aH_E)^{1/2}$ determined by the weak intraplanar anisotropy (for EuTe, for example, $H_\alpha\approx 10$ Oe, Ref. 21) and in fields $(2H_aH_E)^{1/2}\ll H\ll 2H_E$ the RS probability as a function of the field changes and becomes $\propto H$. In other words, the RS is enhanced when long-wave acoustic magnons are created in this region of external field; this agreed with the exchange enhancement, noted in Ref. 21, of RS in noncollinear orthoferrites. For arbitrary [but stronger than $(2H_aH_E)^{1/2}$] fields, the matrix element of Stokes RS with excitation of acoustic magnons having $\mathbf{k}\approx\mathbf{Q}\approx 0$ takes the form

$$w_{sc}^{\text{Stokes}}=-4\pi(\omega_{sc}\omega_{in})^{1/2}e_{sc}{}^{i}c_{ij}^{(S)}e_{in}{}^{j}\frac{\varepsilon_S^{(f)}}{\varepsilon_S{}^{(f)}(H)}\left(\frac{g_f}{g}\frac{\varepsilon_S}{\varepsilon_S{}^{(f)}}-\varkappa\right)\times\sin\theta\cos^{1/2}\theta. \qquad (17)$$

It can be seen from (17), and also from (16), that the quantity $\omega_{sc}^{\text{Stokes}}$ is indeed proportional to the difference $g_fI-\varkappa g|I_f|$, contained in the value of the shift of the energies of the excited states as a function of the field (see $\mathscr{E}_{M_fM}$ in Eqs. (14) and (8)). And if $I-|I_f|\sim I$, then the intensity of the exchange RS at its maximum ($H\approx H_E$) becomes comparable with the spin-orbit scattering intensity,[2)] since usually $I_{sz}\sim\lambda_{SO}$ ($\sim$(10–10^2) cm^{-1}, where λ_{SO} is the spin-orbit constant.

In concluding this section, it is important to emphasize that although anisotropic RS is indeed observed in strongly canted AFM, the canting itself is a necessary but not sufficient condition for the onset of the exchange mechanism. What is important here is not so much this circumstance as the presence of another "shear, or an angle $\beta_{\mathbf{H}}=\theta-\varkappa\theta_f\neq 0$, which appears at $g_fI\neq g\varkappa|I_f|$. The latter, as already mentioned, causes (or can cause) also a shift of the optical-absorption edge on magnetization (if $g_fI=g\varkappa|I_f|$, then, neglecting the spatial dispersion of the electronic excitations, an external field $\mathbf{H}\perp Z$ does not cause a shift of the aforementioned edge in an AFM with EP anisotropy).

THE DIELECTRIC TENSOR

It is known that various magneto-optic effects are described by the magnetic part $\Delta\varepsilon_{ij}^{(S)}(\omega)$ of the dielectric tensor $\varepsilon_{ij}(\omega)$. The explicit form of $\Delta\varepsilon_{ij}^{(S)}(\omega)$ can be established by using only symmetry considerations and expanding $\varepsilon_{ij}(\omega)$ in terms of the components of the ferro- and antiferromagnetism vectors. In this case, a sufficiently reliable estimate of the phenomenological constants of this series calls for calculations that use specific microscopic mechanisms. It is of interest therefore to calculate the tensor $\varepsilon_{ij}(\omega)$ within the framework of the model developed above. This may turn out to be useful also because it makes it possible to extract from magneto-optic data information on the electronic and magnetic parameters of the excited states, on the oscillator strengths, and on the values of the ion-ion interactions. One of them is the resonant interaction contained in the operator (2); in the creation-operator representation (11) it can be written in the form

$$V_{res}^{(f)}=L\sum_{n\alpha,\rho}\sum_{M,M_f,M_f'}d_{M_fM}^{S}(\beta_{\mathbf{H}})d_{M_f'M}^{S}(\beta_{\mathbf{H}})\times B_{n\alpha}{}^{+}(M_f;M)B_{n\alpha+\rho}(M;M_f'), \qquad (18)$$

where L is the constant of this interaction for the nearest neighbors. Expression (18) shows that "mixing" of the spin projections in the processes that involve excitation transfer between the ions is possible, just as in (12), only if $\beta_{\mathbf{H}}\neq 0,\pi$.

Just as any kinetic coefficient, the tensor $\varepsilon_{ij}(\omega)$ can be calculated from a Fourier component of the Green's function, which in this case can be defined as

$$G_{M_fM}^{(\alpha\beta)}(\omega;\mathbf{k})=\langle\!\langle B_{n\alpha}(M;M_f;t)\,|\,B_{m\beta}{}^{+}(M_f;M)\rangle\!\rangle_{\omega;\mathbf{k}}, \qquad (19)$$

where

$$\langle\langle B_{n\alpha}(M;M_f;t)\,|\,B_{m\beta}{}^{+}(M_f;M)\rangle\rangle=-i\theta(t)\,\text{Sp}\,e^{-\mathscr{H}S/k_BT}\times[B_{n\alpha}(M;M_f;t),B_{m\beta}{}^{+}(M_f;M)],$$

$$B_{n\alpha}(M;M_f;t)=\exp[i(\mathscr{H}_S+\mathscr{H}^{(f)})t]B_{n\alpha}(M;M_f)\exp[-i(\mathscr{H}_S+\mathscr{H}^{(f)})t],$$

$\theta(t)$ is the Heaviside function, and the Hamiltonian $\mathscr{H}^{(f)}$ has been excluded from the statistical operator, for usually $\mathscr{E}\gg k_BT$ for electronic excited terms. A closed system of equations for the functions (19) can be obtained by starting from the standard equations of motion in which a decoupling procedure is carried out.[16] Retaining in this case the mean values of only slowly varying quantities, which include the previously introduced value s of the average spin per site and the mean value of the operator $P_{n\alpha}(M)$ [Eq. (10)], we arrive at the system

$$(\omega-\mathscr{E}_{M_fM})G_{M_fM}^{(\alpha\beta)}(\omega;\mathbf{k})=P(M)\left\{1+L[d_{M_fM}^{S}(\beta_{\mathbf{H}})]^2\gamma(\mathbf{k})\sum_{\alpha'}G_{M_fM}^{(\alpha'\beta)}(\omega;\mathbf{k})\right\} \qquad (20)$$

$$\gamma(\mathbf{k})=\frac{1}{z}\sum_{\rho}e^{i\mathbf{k}\rho},$$

for which, to simplify the expression, account was taken in (18) of only resonant ($M_f' = M_f$) terms (this can be done if $|L| < |I - I_f|$). Equating to zero the determinant of the resultant system, we obtain the eigenenergies

$$\mathscr{E}^{(\pm)}_{M_f M}(\mathbf{k}) = \mathscr{E}_{M_f M} \pm L[d^{S}_{M_f M}(\beta_{\mathbf{H}})]^2 P(M)\gamma(\mathbf{k}) \qquad (21)$$

of the dipole excitations (excitons) of the AFM. Substitution of these expressions in (20) yields corresponding Green's functions $G^{(\pm)}_{M_f M}(\omega;\mathbf{k})$ that have simple poles (21). Using, finally, the known relation between the tensor $\varepsilon_{ij}(\omega)$ and its Green's function (see, e.g., Ref. 22), we get ultimately

$$\varepsilon^{(\pm)}_{ij}(\omega) = 1 + \frac{4\pi\varepsilon}{v\omega}(d_1{}^i \pm d_2{}^i)(d_1{}^j \pm d_2{}^j)$$

$$\times \sum_{M, M_f} [d^{S}_{M_f M}(\beta_{\mathbf{H}})]^2 [G^{(\pm)}_{M_f M}(\omega; \mathbf{Q}) - G^{(\pm)}_{M_f M}(-\omega; -\mathbf{Q})], \qquad (22)$$

where the tensors $\varepsilon^{(\pm)}_{ij}(\omega)$ determine the response of an EP-type AFM to an electromagnetic wave with respective polarization $\mathbf{e} \| \mathbf{d}_1 \pm \mathbf{d}_2$, and v is the volume of the magnetic cell. It can be seen directly from (22) that the tensors $\varepsilon^{(\pm)}_{ij}(\omega)$ contain no diagonal components, i.e., do not "mix" the polarizations. This is as it should be since, as already noted, the very equality $\varepsilon^{(\pm)}_{ij}(\omega) = 0$ at $i \neq j$ was the result of the tendency to consider optical effects due only to exchange interaction, and therefore to neglect the orbital structure of the electronic term and hence of the dependence of the operator (12) on the circular polarization of the light. This means in turn that expression (22) does not contain terms that describe magneto-anisotropic phenomena (known to be due to spin-orbit interaction[3]), and corresponds to the refractive indices in the transparency region, where the exciton damping is negligible.

We consider now the magnetic contributions to the refractive index n of a cubic AFM, using the standard definition $n^2 = \mathrm{Re}\varepsilon(\omega)$ (Ref. 22), where $\varepsilon(\omega) = \varepsilon^{(\pm)}_{ii}(\omega)$. Recognizing then that in the tranparency region we have $\mathscr{E} - \omega \gg M\varepsilon_S, M_f\varepsilon^f_S(\mathbf{H}), L$, and carrying out the expansion accurate to terms of first order, we arrive at the expression

$$n^2 = n_0^2 + 2n_0\Delta n_S, \qquad n_0^2 = 1 - \frac{16\pi\mathscr{E}}{v}\frac{d^2}{\omega^2 - \mathscr{E}^2},$$

$$\Delta n_S = \frac{n_0^2 - 1}{n_0}\frac{\mathscr{E}}{\mathscr{E}^2 - \omega^2}\sum_{M}\Big\{[\varepsilon_S M - \varepsilon_S{}^{(f)}(\mathbf{H}) \times \sum_{M_f} M_f [d^{S}_{M_f M}(\beta_{\mathbf{H}})]^2 + L\sum_{M_f}[d^{S}_{M_f M}(\beta_{\mathbf{H}})]^4 P(M)\Big\}P(M), \qquad (23)$$

where Δn_S determines the sought-for magnetic contribution which, being isotropic, depends both on the temperature and on the field. As to the temperature dependence, it can be seen from expressions (20)–(23) that it is determined not only by the ion-ion (exchange and resonant) interactions, but also by the change (via population of the spin sublevels of the ground multiplet) of the oscillator strength of the transition, as well as by the specific value of S. For example, for $S = 1/2$ the mean values are $P(\pm 1/2) = 1/2 \pm s$ and in the absence of a field it follows from (23) that $\Delta n_S \sim s^2$ independently of the relation between ε_S, ε^f_S, and L; such a relation was observed in a number of AFM (see Ref. 23 and the bibliography therein) For $S > 1/2$ the temperature dependence of Δn_S can also be obtained by substituting in (23) explicit expressions for $P(M)$. For arbitrary S they are obtained by representing the operators $(S^{\zeta}_{n\alpha})^m (m \leqslant 2S)$ in terms of the operators (10) and using the normalization condition $\Sigma_M P_{n\alpha}(M) = 1$. Direct calculation of the mean values $\langle\{S^{\zeta}_{n\alpha}\}^m\rangle$, and hence or $P(M)$, can be carried out by the method of Tahir-Kheli and ter Haar (see, e.g., Ref. (16). It turns out then that if allowance for the level shift leads to the same dependence of Δn_S on only s_2, the resonant interaction leads both to a similar behavior or to a smoother one, connected with the variation of the multipole spin moments with temperature (for $S = 1$, in particular, these are quadrupole moments, on which Δn_S depends,[10] as well as, for example, the polarizability of rare-earth materials[24] when account is taken of the effects of the crystal field or of one-ion anisotropy).

The behavior of Δn_S in an external field is determined by the elements of the irreducible representation of the rotation group, or by the functions $d^{S}_{M_f M}(\beta_{\mathbf{H}})$ whose explicit form depends on S. It is easy to verify, however, that in the exchange approximation, for example (when the spatial dispersion, i.e., the width of the exciton band, is neglected) the magnetic contribution to the refractive index is $\Delta n_S \sim \cos\beta_{\mathbf{H}}$ regardless of the value of S. Taking Eq. (7) into account, we arrive at the simple expression ($T \ll T_N$)

$$\Delta n_S = \frac{n_0^2 - 1}{n_0}\frac{\mathscr{E}}{\mathscr{E}^2 - \omega^2} \times \left\{\varepsilon_S - \varepsilon_S{}^{(f)}\left[\varkappa + 2\left(\frac{g_f}{g}\frac{I}{|I_f|} - \varkappa\right)\cos^2\theta\right]\right\}, \qquad (24)$$

from which it is seen directly that the isotropic magnetic contribution is determined by the square of the magnetic moment induced by the external field. The latter corresponds in fact to the phenomenological expression for the magnetic part of the tensor $\varepsilon_{ij}(\omega)$ when $\Delta\varepsilon^{(S)}_{ij}(\omega)$ is given by an expansion in the ferro- and antiferromagnetism vectors.[1,3] Although it was noted in Ref. 1 that the isotropic contribution has a quadratic dependence on the magnetization, it should be noted that allowance for the spatial dispersion of the excitons in (23) leads to a dependence of Δn_S on higher even powers of the magnetic field. It can be stated that the dependence of Δn_S on $\mathbf{H}$ in an AFM with EP anisotropy is determined completely by parameters pertaining to the excited states, while at $g_f I = \varkappa g|I_f|$ it is determined only by the collective nature of the electronic excitations. From (24) we can estimate the total change of the refractive index of an AFM when the crystal is magnetized to saturation; for exchange fields (10^2–10^3) kOe and transition energies (10^4–10^5) cm^{-1} the value of Δn_S can range from 10^{-3} to 10^{-1} in accord with the observed values[23] ($\Delta n_S = 5 \cdot 10^{-2}$ in EuTe).

CONCLUSION

Thus, the exchange RS mechanism and the isotropic magnetic contribution to the refractive index of AFM demonstrate the important role of exchange interaction between excited and unexcited ions (this can include, in principle, also exchange between d-electron spins and localized f-mo-

ments in magnetic semiconductors, although a separate analysis is required for the latter problem). The difference of the parameters corresponding to this interaction from exchange in the ground state produces in a canted AFM deformation of the spin configuration and, as a result, scattering by spin waves without change of the polarization of the light. Of course, in real magnetic system the exchange RS mechanism competes with the spin-orbit mechanism one should apparently speak of an increase, with increase of H, of the fraction of scattered light that has the same polarization of the incident one. It must be borne in mind here that the direction of **H** is such that the quantization axes of the spins differ from those of the orbital momentum, the diagonal components of the tensor of the one-magnon RS can in principle become different from zero also in the mechanism of the **LS** spin-orbit interaction, which will contain mixed terms (L^iS^j, $i \neq j$) in the proper coordinate frames. If $\mathbf{H} \perp Z$, however, this does not take place in an AFM with EP anisotropy, and the exchange mechanism should be most clearly pronounced. Interest attaches therefore to an experimental study of RS in AFM of the EP type in a strong field perpendicular to the symmetry axis, with an aim of observing the "non-rotated" component scattered by the spin waves. It is also obvious that the deformation of the spin structure of the excited state should be manifest also in other optical phenomena, such as the field dependence and magnitude of the Davydov splitting, in magneto-optic anisotropy, etc.

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[1)]Note that exchange interaction of orbitally excited and unexcited ions is known to be the cause of many effects. Thus, resonant exchange interaction determines the dispersion of magnetic excitons and Davydov splitting of the corresponding magnetodipole absorption lines in AFM.[9,10] On the other hand, the magnetic contributions to the refractive index of even collinear magnets are due to a considerable degree to the fact that the static exchanges in the ground and dipole-excited states are unequal.[11,12]

[2)]The RS probability in the spin-orbit interaction mechanism was estimated in many papers.[5,6,8] The difficulty is that there is no reliable information on the quantities contained in the definition of $c_{ij}^{(S)}$ (dipole moments, oscillator strengths, transition strengths, virtual-state energies). Thus, for an infrared or optical photons, dipole moments ~ 1 a.u., and transition energies $(10^4$–$10^5)$ cm^{-1} the scattering probability (or the exctinction coefficient) per unit interval of solid angle and frequency can take on values from 10^{-12} cm^{-1} (Ref. 6) to 10^{-5} cm^{-1} (Ref. 8); experiment yields $\sim(10^{-10}$–$10^{-11})$ cm^{-1} (Refs. 8 and 9). According to Ref. 1, for EuTe this value is $(10^{-8}$–$10^{-9})$ cm^{-1}, and the exchange RS begins to be observed in fields ~ 20 kOe ($2H_E = 72$ kOe, Refs. 1 and 2).

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