Free and self-trapped excitons in antiferromagnetic insulators

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A study is made of the photoexcitation of ions in antiferromagnetic insulators (AFIs) with regard to how variation of the exchange interaction between a magnetic ion and its environment affects the shape of the optical absorption spectrum and the diffusion of excitations. It is shown that for weak relaxation within a multiplet, absorption of light in the AFI can give rise to self-trapped as well as free excitons; self-trapping occurs by way of an inhomogeneity in the distribution of spins which makes up the spin cloud surrounding the photoexcited ion. In the case of strong relaxation within the multiplet, the self-trapped exciton moves through the crystal diffusively; the diffusion coefficient depends on the external magnetic field and the direction of exciton propagation.

INTRODUCTION

As a rule, in antiferrogmagnetic insulators (AFIs) containing ions from the iron group, optical excitation is accompanied by a change in the multiplicity of the electronic state of the $3d$ electrons,[1,2] In this case, migration of the quantum excitation to a neighboring magnetic ion can occur, due to the exchange resonant interaction (ERA) between $3d$-electrons.[3–5] Particularly clear evidence of the ERA is found in the intersublattice Davydov splitting of the exciton lines, which provides direct evidence of the Frenkel-like nature of optical excitations in AFIs.[6–8] As a consequence of its exchange nature, the magnitude of the ERA depends on the mutual alignments of the magnetic ion spins. For the ground-state electronic configuration the spin directions in the case of weak single-ion anisotropy coincide with the directions of the sublattice magnetizations. In what follows, we will choose the spin quantization axis (SQA) along these directions. As was shown in Ref. 9, the quantity $L^{f}_{\mathbf{n}\alpha;\mathbf{m}\beta}$ of the ERA for two ions at $\mathbf{n}\alpha$ and $\mathbf{m}\beta$ in the magnetic sublattices α and β as a function of the angle $\theta_\alpha - \theta_\beta$ between the SQA of the ions at $\mathbf{n}\alpha$ and $\mathbf{m}\beta$ is given by the expression

$$L^{f}_{\mathbf{n}\alpha;\mathbf{m}\beta}=\mathscr{M}^{f}_{\mathbf{n}\alpha;\mathbf{m}\beta}\cos^2\frac{\theta_\alpha-\theta_\beta}{2}, \qquad (1)$$

in which $\mathscr{M}^{f}_{\mathbf{n}\alpha;\mathbf{m}\beta}$ is a parameter which determines the nature of the f-optical excitations in a specific AFI. It is clear from Eq. (1) that the ERA is maximal for parallel spin alignments ($\theta_\alpha - \theta_\beta = 0$) and zero for antiparallel spin alignments ($\theta_\alpha - \theta_\beta = \pi$). Because an external magnetic field H_0 can give rise to noncollinearity of the spins for $\alpha \neq \beta$ sublattices in the case of weak single-ion anisotropy, the magnitude of the ERA (Eq. 1) is itself a function of the external field. The dependence (1) explains the Davydov splitting induced by the magnetic field in weakly-anisotropic AFIs,[3–5] and also the behavior of the diffusion coefficient of self-trapped excitons in the quasi-one-dimensional AFI $NaMnCl_3$ (Ref. 10).

At the present time, the theory of excitonic excitations in AFIs (see Refs. 3 and 4 for an exposition of this) is based on the assumption that the SQAs of a magnetic ion coincide for the ground and excited states.[1)] It follows from recent experiments, however, that such a picture is not valid for every excitation. Thus, Eremenko and co-workers have shown that for excitation of the Mn^{2+} ion to the state $f = {}^4T_1({}^4P)$ in the crystals MnF_2 (Ref. 11) and $NaMnCl_3$ (Ref. 12) the direction of the spin of the photoexcited ion (PI) can vary, both because of the appearance of strong single-ion anisotropy and also because of the variation in the magnitude of the exchange interactions with the neighbors. As a consequence of the second of these reasons, it is also possible to have an additional "tilt" of the ion spins which neighbor the PI.[12,13] The presence of such a "tilt" leads to the formation around the PI of a spin cloud, which hinders the hopping of the excitations and thereby makes possible the self-trapping of an excitation on a specific PI. This situation recalls the case which arises in nonmagnetic crystals when a phonon cloud appears around a photoexcited molecule[14,15] or an atom.[16] Recently, it was shown by Rashba and Toyozawa (see, e.g., Refs. 17, 18) that in a nonmagnetic crystal with a strong electron-phonon coupling it is possible for self-trapped excitons as well as free excitons to form; the characteristic peculiarity of the former is its strong localization in the strain field formed by the optical excitation. The narrower the exciton band at the original equilibrium positions of the nuclei and the weaker the exciton-phonon coupling, the stronger the self-trapping will be. For an AFI with Frenkel-type excitons, in addition to the vibrational degrees of freedom, the spin degrees of freedom also play an important role in dynamic processes. The subject of the present article is an investigation of the role of the latter in the process of formation of excitonic states in AFI, and also in the migration of self-trapped optical excitations. We will assume that single-ion anisotropy in the ground and optically excited multiplet is weak.

RESONANCE INTERACTION BETWEEN MAGNETIC IONS

Within the framework of the molecular field (MF) approximation[19,20], let us assume the eigenfunctions $|O_{\mathbf{n}\alpha}(M_{\mathbf{n}\alpha})\rangle$, $|f_{\mathbf{n}\alpha}(M^{f}_{\mathbf{n}\alpha})\rangle$ of a magnetic ion at lattice site $\mathbf{n}$ of sublattice α in its ground and f-excited configuration are known. Here, the spin projections $M_{\mathbf{n}\alpha}$ and $M^{f}_{\mathbf{n}\alpha}$ are defined relative to the SQA ζ_α and $\zeta^{f}_{\mathbf{n}\alpha}$, which usually do not coincide for the ground and excited multiplets (because the directions and magnitudes of the exchange fields acting on a particular ion need not coincide[13]). In the presence of the $\mathbf{n}\alpha$th PI there is also a change in the exchange field which acts on the spins of the ions $j(\mathbf{n}\alpha)$ adjacent to $\mathbf{n}\alpha$. The corresponding wave functions $|\tilde{O}_{j(\mathbf{n}\alpha)}(M_{j(\mathbf{n}\alpha)})\rangle$ are defined relative to the SQA $\zeta_{j(\mathbf{n}\alpha)}$. Because of the weak single-ion anisot-

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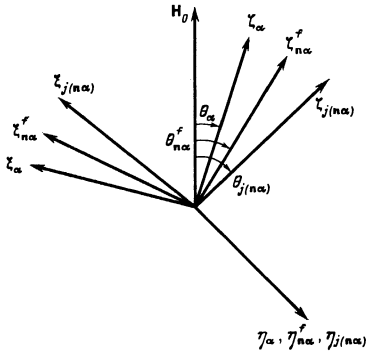

FIG. 1. Directions of the intrinsic SQAs of ion $n\alpha$ while in the ground ($\zeta\alpha$) and optically-excited ($\zeta^f_{n\alpha}$) electronic states; $\zeta_{j(n\alpha)}$ is the SQA of ion j in the ground electronic state near the PI $n\alpha$.

ropy, the SQAs $\zeta_\alpha, \zeta^f_{n\alpha}, \zeta_{j(n\alpha)}$ all lie in a single plane and are determined by the angles θ_α, $\theta^f_{n\alpha}$, $\theta_{j(n\alpha)}$ relative to the external field $\mathbf{H}_0$ (see Fig. 1). For an explicit expression for these angles in the case of a two-sublattice AFI, see Refs. 12, 13. Within the framework of the MF approximation, the wave functions of the AFI in the absence of perturbations and in the presence of a PI at $n\alpha$ have the form

$$|O\{M_j\}\rangle=\prod_j |O_j(M_j)\rangle, \tag{2}$$

$$|F_{n\alpha}(M_{n\alpha}{}^f,\{M_{j(n\alpha)}\})\rangle=|f_{n\alpha}(M_{n\alpha}{}^f)\rangle\prod_{j(\neq n\alpha)}|\bar{O}_{j(n\alpha)}(M_{j(n\alpha)})\rangle. \tag{3}$$

For convenience in writing, let us choose the sign of the Zeeman energy so that the maximum possible spin projection corresponds to the smallest energy. Then wave function (2) with $M_j = S$ will correspond to the ground state of the AFI at $T=0$ K, while function (3) with $M_{j(n\alpha)} = S$ and $M^f_{n\alpha} = S-1$ corresponds to the lowest spin-forbidden excited energy level if the sign of the exchange field for the photoexcited ion at $n\alpha$ does not change; likewise, the function (3) with $M_{j(n\alpha)} = S$ and $M^f_{n\alpha} = -(S-1)$ corresponds to the lowest spin-forbidden level if the sign of the exchange field does change.

Because of the translation symmetry of the crystal and the equivalence of the magnetic sublattices, the state described by wave function (3) is degenerate. The degeneracy is lifted by the ERA, leading to quantum transfer of the electronic excitation f from ion $n\alpha$ to ion $n\beta$. The corresponding matrix element

$$L^f_{n\alpha;m\beta}=\langle F_{m\beta}|V_{n\alpha;m\beta}|F_{n\alpha}\rangle,$$

where $|F_{n\alpha}\rangle$, $|F_{m\beta}\rangle$ denote function (3), is calculated using the ERA operator

$$V_{n\alpha;m\beta}=2\mathcal{M}^f_{n\alpha;m\beta}\hat{\tau}_{n\alpha}\hat{\sigma}_{m\beta},$$

in which the operator $\hat{\sigma}$ generates a transition within the ion from the ground-state multiplet to a state of the excited multiplet, while the operator $\hat{\tau}$ mediates the reverse; then

$$\begin{aligned}\langle f(M')|\hat{\sigma}^\mp|O(M)\rangle&=\langle O(M)|\hat{\tau}^\pm|f(M')\rangle\\&=\pm\delta_{M',M\mp1}\left[\frac{(S\pm M)(S\pm M-1)}{2S(2S-1)}\right]^{1/2},\\\langle f(M')|\hat{\sigma}^\xi|O(M)\rangle&=\langle O(M)|\hat{\tau}^\xi|f(M')\rangle\\&=-\delta_{M',M}\left[\frac{(S-M)(S+M)}{2S(2S-1)}\right]^{1/2}.\end{aligned} \tag{4}$$

Here $\hat{\sigma}^\pm\equiv\hat{\sigma}^\xi_j\pm i\hat{\sigma}^\eta_j$, $\hat{\sigma}^\zeta\equiv\hat{\sigma}^\zeta_j$, where $\xi=\xi_j$, $\eta=\eta_j$, $\zeta=\zeta_j$ is a system of coordinates in which ζ_j is the SQA for the jth ion (one of the three shown in Fig. 1). In the process of finding $L^f_{n\alpha;m\beta}$, we encounter the matrix elements

$$\langle\bar{O}_{n\alpha(m\beta)}(M_{n\alpha(m\beta)})|\hat{\tau}_{n\alpha}|f_{n\alpha}(M_{n\alpha}{}^f)\rangle,$$

$$\langle f_{m\beta}(M_{m\beta}{}^f)|\hat{\sigma}_{m\beta}|\bar{O}_{m\beta(n\alpha)}(M_{m\beta(n\alpha)})\rangle,$$

in order to calculate them, we must first transform to a common SQA before using Eq. (4). Let us choose for the SQA $\zeta_{n\alpha(m\beta)}$ for the first matrix element and $\zeta_{m\beta(n\alpha)}$ for the second. Then

$$\begin{aligned}&\langle\bar{O}_{n\alpha(m\beta)}(M_{n\alpha(m\beta)})|\hat{\tau}_{n\alpha}|f_{n\alpha}(M^f_{n\alpha})\rangle\\&=\sum_{M_1{}^f}d^{s-1}_{M^f_{n\alpha}M_1{}^f}(\theta^f_{n\alpha}-\theta_{n\alpha(m\beta)})\,\mathbf{K}_{n\alpha}(M_1{}^f\to M_{n\alpha(m\beta)}),\end{aligned} \tag{5}$$

where $d^s_{MM'}$ is the Wigner transition matrix for spin S from one coordinate system to the other, and

$$\mathbf{K}_{n\alpha}(M_1{}^f\to M_{n\alpha(m\beta)})=\langle\bar{O}_{n\alpha(m\beta)}(M_{n\alpha(m\beta)})|\hat{\tau}_{n\alpha}|f_{n\alpha}(M_1{}^f)\rangle, \tag{6}$$

corresponding to Eq. (4) we have

$$\begin{aligned}&\mathbf{K}_{n\alpha}(M^f\to M)\\&=\frac{1}{(2S(2S-1))^{1/2}}\Big[-\delta_{M',M}(S^2-M^2)^{1/2}\mathbf{n}_\zeta+\frac{1}{2}\delta_{M',M-1}\\&\quad\cdot[(S+M)(S+M-1)]^{1/2}(\mathbf{n}_\xi-i\mathbf{n}_\eta)\\&\quad-\frac{1}{2}\delta_{M',M+1}[(S-M)(S-M-1)]^{1/2}(\mathbf{n}_\xi+i\mathbf{n}_\eta)\Big].\end{aligned} \tag{7}$$

In Eq. (7), $\mathbf{n}_\xi$, $\mathbf{n}_\eta$, $\mathbf{n}_\zeta$ are unit vectors along the axes of the coordinate system $\xi_{n\alpha(m\beta)}$ $\eta_{n\alpha(m\beta)}$ $\zeta_{n\alpha(m\beta)}$. Equations analogous to (5)–(7) are also obtained for the second matrix element mentioned above. Using these equations, we obtain

$$\begin{aligned}L^f_{n\alpha;m\beta}\equiv L^f_{n\alpha;m\beta}&(M^f_{n\alpha}\to M_{n\alpha(m\beta)};\ M_{m\beta(n\alpha)}\\&\to M^f_{m\beta};\{M_{j(n\alpha)}\to M_{j(m\beta)}\})\\&=2\mathcal{M}^f_{n\alpha,m\beta}\prod_{j(\neq n\alpha,\,m\beta)}d^s_{M_{j(m\beta)}M_{j(n\alpha)}}(\theta_{j(m\beta)}\\&\quad-\theta_{j(n\alpha)})\sum_{M_1{}^fM_2{}^f}d^{s-1}_{M^f_{n\alpha}M_1{}^f}(\theta^f_{n\alpha}-\theta_{n\alpha(m\beta)})\\&\quad\cdot d^{s-1}_{M^f_{m\beta}M_2{}^f}(\theta^f_{m\beta}-\theta_{m\beta(n\alpha)})\,\mathbf{K}_{n\alpha}(M_1{}^f\to M_{n\alpha(m\beta)})\\&\quad\cdot\mathbf{K}^*_{m\beta}(M_2{}^f\to M_{m\beta(n\alpha)}).\end{aligned} \tag{8}$$

It follows from Eq. (8) that the effectiveness of the quantum hopping depends both on the magnitude of the deivation of the SQA $\zeta^f_{n\alpha}$, $\zeta_{j(n\alpha)}$ from the direction ζ_α of the magnetizations of the sublattices (the factors $d^s_{MM'}$ and $d^{s-1}_{MM'}$) and on the mutual orientation of the SQAs $\zeta_{n\alpha(m\beta)}$ and $\zeta_{m\beta(n\alpha)}$ (the product $\mathbf{K}_{n\alpha}\cdot\mathbf{K}_{m\beta*}$, Eq. (8) also makes evident the fact

that an electronic transition within a specific ion is correlated with transitions between spin components of multiplets on surrounding ions which remain in their ground state configuration while changing their SQA. In other words, there occurs a dressing of the optical excitation within a given ion by the spin clouds of the other ions. This is reminiscent of exciton self-trapping caused by phonons.[14-18] However, here the role of the nuclear degrees of freedom is played by the spin degrees of freedom. The situation somewhat resembles the process of creation of a spin cloud around a moving electron in magnetic semiconductors during the formation of a spin-polaron or magnetic polaron (see, e.g., Refs. 24, 25). The difference here is that the role of the electron is played by a currentless spin-forbidden optical excitation.

At $T=0$ K the magnetic ions have their maximal spin projections along their intrinsic SQAs, i.e.,

$$M_{\mathbf{n}\alpha(\mathbf{m}\beta)}=M_{\mathbf{m}\beta(\mathbf{n}\alpha)}=M_{j(\mathbf{n}\alpha)}=M_{j(\mathbf{m}\beta)}=S.$$

Therefore, the matrix element of the ERA corresponding to the lowest excitation ($M^f_{\mathbf{n}\alpha}=M^f_{\mathbf{m}\beta}=S-1$), according to (8), (7), has the form

$$L^f_{\mathbf{n}\alpha;\mathbf{m}\beta}=L^f_{\mathbf{n}\alpha;\mathbf{m}\beta}((S-1)\to S;\ S\to(S-1);\ \{S\to S\})$$

$$=\mathscr{M}^f_{\mathbf{n}\alpha;\mathbf{m}\beta}\cos^2\left(\frac{\theta_{\mathbf{n}\alpha(\mathbf{m}\beta)}-\theta_{\mathbf{m}\beta(\mathbf{n}\alpha)}}{2}\right)\cos^{2(s-1)}\left(\frac{\theta^f_{\mathbf{n}\alpha}-\theta_{\mathbf{n}\alpha(\mathbf{m}\beta)}}{2}\right)$$

$$\cdot\cos^{2(s-1)}\left(\frac{\theta_{\mathbf{m}\beta}{}^f-\theta_{\mathbf{m}\beta(\mathbf{n}\alpha)}}{2}\right)\prod_{j(\neq\mathbf{n}\alpha,\mathbf{m}\beta)}\cos^{2s}\left(\frac{\theta_{j(\mathbf{m}\beta)}-\theta_{j(\mathbf{n}\alpha)}}{2}\right). \tag{9}$$

Eq. (9) generalizes the dependence on spin noncollinearity of the matrix element (1) for resonant transfer of spin-forbidden optical excitations of an AFI, and coincides with (1) in the absence of any deviation of the SQA from the direction of ζ_α, i.e., in the case

$$\theta_{\mathbf{n}\alpha(\mathbf{m}\beta)}=\theta_{\mathbf{n}\alpha}{}^f=\theta_\alpha,\quad \theta_{\mathbf{m}\beta(\mathbf{n}\alpha)}=\theta_{\mathbf{m}\beta}{}^f=\theta_\beta,\quad \theta_{j(\mathbf{m}\beta)}=\theta_{j(\mathbf{n}\alpha)}.$$

FORMATION OF A SPIN-FORBIDDEN EXCITON BAND

For definiteness, let us investigate a two-sublattice weakly-anisotropic AFI in the noncollinear phase for $H_0>H_{cr}$, where H_{cr} is the field of the spin-flop phase. Because for $H_0>H_{cr}$ the SQAs for the various sublattices are aligned symmetrically relative to the external field $\mathbf{H}_0$, we introduce the notation

$$\theta_\alpha=(-1)^{\alpha+1}\tilde\theta,\quad \theta_{\mathbf{n}\alpha}{}^f=(-1)^{\alpha-1}\theta^f,\quad \theta_{j(\mathbf{n}\alpha)}=(-1)^\alpha\tilde\theta. \tag{10}$$

The last equation in (10) is written for the ions $j(\mathbf{n}\alpha)$ which are nearest neighbors of $\mathbf{n}\alpha$ and belong to the opposite sublattice. This limitation on the ions $j(\mathbf{n}\alpha)$ will also be employed below. Within this framework, following Refs. 21 and 13 and using the MF approximation, let us write down the Hamiltonian for an AFI with the $\mathbf{n}\alpha$th ion excited:

$$\mathscr{H}_{\mathbf{n}\alpha}{}^f=E^f(\tilde\theta,\theta^f)-\mu_B g^f H^f\hat S^{f\xi}_{\mathbf{n}\alpha}-\mu_B g_0\tilde H\sum_j\hat S^{\xi}_{j(\mathbf{n}\alpha)}. \tag{11}$$

Here,

$$E^f(\tilde\theta,\theta^f)\equiv E^f-Z\varkappa|I^f|\sigma^f\tilde\sigma S(S-1)\cos(\tilde\theta+\theta^f)$$

is the energy position of the center of gravity of the excited multiplet consisting of a constant part E^f which depends on the external field H_0 (through the angles θ and θ^f), and

$$H^f\equiv H_0\cos\theta^f-\frac{S}{S-1}H_E{}^f\tilde\sigma\cos(\theta+\tilde\theta), \tag{12}$$

$$\tilde H\equiv H_0\cos\tilde\theta-\frac{1}{Z}\varkappa\xi^f H_E{}^f\tilde\sigma\cos(\tilde\theta+\theta^f)-\frac{Z-1}{Z}H_E\sigma\cos(\theta+\tilde\theta) \tag{13}$$

are effective magnetic fields acting on the PI and the ions around it. In the expressions above, σ^f, $\tilde\sigma$ and σ are the specific magnetizations of the PI, the neighboring ion and the ion with fixed SQA; g_0, g^f are g-factors, μ_β is the Bohr magneton, Z is the number of nearest neighbors from the opposite sublattice,

$$H_E\equiv SZI/\mu_B g_0,\quad H_E{}^f=SZ|I^f|/\mu_B g^f$$

are exchange fields acting on the spin of the ion in the ground and excited electronic configurations, $I>0$ is the exchange parameter for the magnetic interaction of an ion with its neighbor, I^f is a similar parameter in the case where one of the ions is photoexcited ($\varkappa\equiv\operatorname{sign} I^f$),

$$\xi^f\equiv(S-1)g^f\sigma^f/Sg_0\tilde\sigma.$$

In Eq. (11), $\hat s_{\mathbf{n}\alpha}{}^{f\xi}$ is the spin projection operator on the axis $\zeta^f_{\mathbf{n}\alpha}$, while $\hat s^\xi_{j(\mathbf{n}\alpha)}$ is the same for a neighboring ion, only taken around the axis $\zeta_{j(\mathbf{n}\alpha)}$.

The wave functions (3) are eigenfunctions for the operator (11) and serve as basis functions for finding the exciton states of the AFI. In order to find the exciton spectrum, we add to (11) the resonance interaction and represent the Hamiltonian for an optically excited state of the AFI in the form

$$\mathscr{H}^f=\sum_{\mathbf{n}\alpha}\langle F_{\mathbf{n}\alpha}|\mathscr{H}_{\mathbf{n}\alpha}{}^f|F_{\mathbf{n}\alpha}\rangle|F_{\mathbf{n}\alpha}\rangle\langle F_{\mathbf{n}\alpha}|$$

$$+\frac{1}{2}\sum_{\mathbf{n}\alpha,\mathbf{m}\beta}L^f_{\mathbf{n}\alpha;\mathbf{m}\beta}|F_{\mathbf{m}\beta}\rangle\langle F_{\mathbf{n}\alpha}|. \tag{14}$$

Because $|F_{\mathbf{n}\alpha}\rangle$ is the state (3) and $L^f_{\mathbf{n}\alpha;\mathbf{m}\beta}$ is defined by Eq. (8), in (14) it is understood that a summation is to be taken over all spin projections of the ground and excited multiplets. During the interaction of the AFI with light, a transition occurs between (2) and (3); the absorption coefficient of light is proportional to the imaginary part of the Fourier transform of the Green's function (see, e.g., Refs. 1, 3–5) for the transition operators $B^+_{\mathbf{n}\alpha}(F)\equiv|F_{\mathbf{n}\alpha}\rangle\langle0|$, $B_{\mathbf{n}\alpha}(F)\equiv|0\rangle\langle F_{\mathbf{n}\alpha}|$.

From here, the averaging is carried out using the density matrix ρ_0 of the ground electronic configuration of the AFI. In the MF approximation, $\rho_0=\Pi_j W(M_j)$, where

$$W(M_j)=\exp(-\hbar\omega^0 M_j/k_BT)/\operatorname{Sp}\exp(-\hbar\omega^0\hat S^\xi_j/k_BT)$$

is the occupation of the M th energy level of the ground-state multiplet of the jth ion, $\hbar\omega^0=\mu_B g_0 H_E\sigma$ is the difference between its nearest energy levels for $2H_E\sigma>H_0>H_{cr}$. The poles of this Green's function give the spectrum of the optical excitations of the AFI. As an example, we present the exciton spectrum corresponding to the transition

$$|O_{\mathbf{n}\alpha}(M)\rangle\to|F_{\mathbf{n}\alpha}(M^f)\rangle$$

on the ion $\mathbf{n}\alpha$ with energy

$$\Delta\varepsilon(M\to M^f)=\Delta\varepsilon^f+\hbar\omega^f M^f-\hbar\omega^0 M,$$

where

$$\hbar\omega^f\equiv\mu_B g^f H^f,\quad \Delta\varepsilon^f\equiv E^f(\tilde\theta,\theta^f)-E_0(\theta),$$

and

$$E_0(\theta)=-ZIS^2\sigma^2\cos 2\theta$$

is the position of the center of gravity of the ground-state multiplet. In the two-sublattice AFI, for $H_0>H_{cr}$ the excitation energy of an ion $\Delta\varepsilon(M\to M^f)$ is split into two exciton bands $E_{1,2}(\mathbf{k},f;M\to M^f)$, characterized by the vector $\mathbf{k}$ and the transition type f. Limiting ourselves for simplicity to the case where only the PI has an inclined SQA, i.e., setting $\tilde\theta=\theta$, $\tilde\sigma=\sigma$, we have

$$E_{1,2}(\mathbf{k},f;M\to M^f)=\Delta\varepsilon^f(M\to M^f)$$
$$+W(M)[Z_1L_1^f(M,M^f)\gamma_1(\mathbf{k})\pm Z|L^f(M;M^f)|\gamma(\mathbf{k})],\quad(15)$$

where Z_1 is the number of nearest neighbors from the same sublattice, and

$$\gamma_1(\mathbf{k})=\frac{1}{Z_1}\sum_{\delta_1}\exp(\delta_1\mathbf{k}),\quad \gamma(\mathbf{k})=\frac{1}{Z}\sum_{\delta}\exp(\delta\mathbf{k})\quad(16)$$

are the geometrical factors for the exciton bands [the summation in (16) ranges over nearest neighbors]. Physically, the factor $W(M)$ reflects the fact that in order for an excitation to undergo an iso-energetic resonance hop from one unexcited ion to another, the latter must have a spin projection of M; the probability that this is the case is precisely $W(M)$. The expressions for $L_1^f(M;M^f)$ and $L^f(M;M^f)$ are obtained from (8), taking into account (7) with $\alpha=\beta$ and $\alpha\neq\beta$, respectively. Because in our case $\theta_{j(\mathbf{m}\beta)}=\theta_{j(\mathbf{n}\alpha)}$, $\theta_{\mathbf{n}\alpha(\mathbf{m}B)}=\theta_\alpha$, $M^f_{\mathbf{n}\alpha}=M^f_{\mathbf{m}\beta}=M^f$, $M_{\mathbf{m}\beta(\mathbf{n}\alpha)}=M_{\mathbf{n}\alpha(\mathbf{m}\beta)}=M$, $M_{j(\mathbf{n}\alpha)}=M_{j(\mathbf{m}\beta)}$,

$$L_1^f(M;M^f)=2\mathscr{M}_1^f\sum_{M_1^fM_1^f}d^{s-1}_{M^fM_1^f}(\theta^f-\theta)\,d^{s-1}_{M^fM_2^f}(\theta^f-\theta)\,\mathbf{K}_\alpha(M_1^f\to M)\times\mathbf{K}_\alpha^*(M_2^f\to M),\quad(17)$$

$$L^f(M;M^f)=2\mathscr{M}^f\sum_{M_1^fM_2^f}d^{s-1}_{M^fM_1^f}(\theta^f-\theta)\,d^{s-1}_{M^fM_2^f}(-\theta^f+\theta)\,\mathbf{K}_1(M_1^f\to M)\times\mathbf{K}_2^*(M_2^f\to M),$$

where $\mathscr{M}_1^f$, $\mathscr{M}^f$ are the quantities $\mathscr{M}^f_{\mathbf{n}\alpha,\mathbf{m}\beta}$ for nearest neighbors when $\alpha=\beta$ and $\alpha\neq\beta$, while $\mathbf{K}_\alpha(M^f\to M)$ is given by Eq. (7) with unit vectors directed along the axes $\xi_\alpha,\eta_\alpha,\zeta_\alpha$ (see Fig. 1). In the absence of a tilt in the SQA of the PI, when $\theta^f=\theta$ we have $d^s_{MM'}(0)=\delta_{MM'}$, and thus $M_1^f=M_2^f=M^f$. Consequently, in (17)

$$\mathbf{K}_1(M^f\to M)\,\mathbf{K}_2^*(M^f\to M)=\frac{1}{2S(2S-1)}\Big\{(S^2-M^2)\delta_{M',M}\cos 2\theta+\frac{1}{2}[(S+M)(S+M-1)\delta_{M',M-1}+(S-M)(S-M-1)\delta_{M',M+1}]\cos^2\theta\Big\},\quad(18)$$

here $\cos\theta=H_0/2H_E\sigma$ (the expression for $\mathbf{K}_\alpha(M^f\to M)\mathbf{K}_\alpha^*(M^f\to M)$ is obtained from (18) formally by setting $\theta=0$).

The light excites states with energies of a $\mathbf{k}=0$ exciton; in correspondence with (15), (16), the splitting between these states is determined by the expression

$$\Delta E(M\to M^f)=2ZW(M)|L^f(M;M^f)|.$$

At low temperatures, when only the lowest level of the ground state multiplet $M=S$ is occupied, and hence $W(M)=\delta_{M,S}$, we obtain from (18) and (17) for $\theta^f=\theta$ the simple expression

$$L^f(S;S-1)=\mathscr{M}^f\cos^2\theta.$$

The corresponding splitting

$$\Delta E(S\to S-1)=2Z|\mathscr{M}^f|\cos^2\theta,$$

which appears as a result of the magnetic field induced spin noncollinearity, was observed in $RbMnF_3$ by Eremenko *et al.*[8] and was explained in Ref. 9 on the basis of the exciton model. Equations (15)–(18) generalize the results of exciton theory to the case of $T\neq 0$ K, when, e.g., there appears a Davydov splitting for $M^f=M\neq S,\ -S$ in agreement with (18), (17) even in a collinear AFI (formally, when $\theta=\pi/2$). For $\theta^f\neq\theta$, exciton theory leads to a fundamentally new result; exciton dispersion along the same sublattice begins also to depend on the magnetic field [as a consequence of the dependence of the parameter $L_1^f(M;M^f)$ on the angle $\theta^f-\theta$ through the factor $d^{s-1}_{MM'}(\theta^f-\theta)$]. One can see this intuitively at $T=0$ K; since $W(M)=\delta_{M,S}$, we obtain from (17) and (7), taking into account the explicit form of $d^{s-1}_{MM'}$,

$$L_1^f(M;M^f)=\delta_{M,S}\delta_{M',S-1}\mathscr{M}_1^f\cos^{4(s-1)}\left(\frac{\theta^f-\theta}{2}\right),\quad(19)$$

$$L^f(M;M^f)=\delta_{M,S}\delta_{M',S-1}\mathscr{M}^f\cos^2\theta\cos^{4(s-1)}\left(\frac{\theta^f-\theta}{2}\right).\quad(20)$$

According to Ref. 13 (see also Ref. 23), the dependence of the angle θ^f on the field H_0 when only the PI has an inclined SQA is given by the expression

$$\sin\theta^f=\frac{\varkappa H_E^f\sigma}{H^{(f)}}\sin\theta,\quad \cos\theta^f=\frac{H_0-\varkappa H_E^f\sigma\cos\theta}{H^{(f)}},\quad(21)$$

where

$$H^{(f)}=[H_0^2+(H_E^f\sigma)^2-2\varkappa H_0H_E^f\sigma\cos\theta]^{1/2}$$

is the effective magnetic field acting on the PI spin in the interval of fields $H_{cr}\leqslant H_0\leqslant 2H_E\sigma$. From (21) it is clear that a nontrivial deviation θ^f from θ arises only in the noncollinear case (in a collinear AFI, when $H_0=0$, $\theta=\pi/2$ we have $\theta^f=\pi/2,\ -\pi/2$ depending on the sign of f^f).

Let us note that the formation of an exciton spectrum in an AFI is possible if the dynamic processes related to the resonance hopping of an excitation from ion to ion, which are characterized by a time $\tau_{res}\sim|L_1^f|^{-1}$, take place faster than the relaxation within a multiplet (characterized by the time τ_{rel}. However, the condition $\tau_{res}\ll\tau_{rel}$ says nothing about whether an additional tilt in the ion spins under photoexcitation is possible or not. Such an additional tilt is possible if the width of the exciton band without a spin tilt of the PI and neighboring ions (for the two-sublattice AFI, the

band is determined by the quantities L_1^f, L^f in (15), if in the expression (17) we set $\theta^f = \theta$) is smaller than the difference in the energies $\Delta\varepsilon a_f$ in (15), taken for $\theta^f \neq \theta$ and $\theta^f = \theta$ (including the spin tilts of the neighboring ions—the difference $E^f(\tilde{\theta},\theta^f) - E^f(\tilde{\theta}=\theta,\theta^f=\theta)$ of the centers of gravity of the excited multiplet). Because for a large photoinduced spin tilt the width of the exciton band is small, self-trapping of the exciton takes place, similar to what occurs in a nonmagnetic crystal because of a photoinduced shift of the nuclei.[17,18]

DIFFUSION COEFFICIENT FOR SELF-TRAPPED EXCITONS IN AN AFI

Let us now investigate an AFI in which the widths of the exciton bands are small, so that the condition $\tau_{\text{rel}} \ll \tau_{\text{res}}$ is fulfilled. A large value of τ_{res} can arise from several causes, e.g., strong exciton-phonon coupling[14] (in which case the quantity $\mathscr{M}^f_{\mathbf{n}\alpha;\mathbf{m}\beta}$ contains the parameter for this coupling) or a large variation in the SQA of the PI and neighboring ions when excited. Assume an excitation lives for a characteristic time τ_{ex}. After the lapse of this time, it can, e.g., decay into luminescence, be caught by a trapping center, or degrade into heat.[26] If $\tau_{\text{ex}} \gg \tau_{\text{res}}$, then by virtue of the condition $\tau_{\text{rel}} \ll \tau_{\text{res}}$, relaxation processes will ensure that at any time interval Δt satisfying the condition $\tau_{\text{rel}} \ll \Delta t \ll \tau_{\text{res}}$, the Boltzmann distribution will be preserved for the level occupation in each of the multiplets. No coherence is involved in the process of excitation hopping from one ion to another for times on the scale Δt; hence, the excitation hopping process in AFI will have a diffusive character.

The probability of incoherent hopping of an excitation from ion $\mathbf{n}\alpha$ to ion $\mathbf{m}\beta$ in a two-sublattice AFI for $H_0 > H_{\text{er}}$ is determined by the expression

$$W^f_{\mathbf{n}\alpha;\,\mathbf{m}\beta} = \frac{2\pi}{\hbar^2} \sum_{\{MM^f\}} |L^f_{\mathbf{n}\alpha;\,\mathbf{m}\beta}(M^f_{\mathbf{n}\alpha} \to M_{\mathbf{n}\alpha(\mathbf{m}\beta)};$$

$$M_{\mathbf{m}\beta(\mathbf{n}\alpha)} \to M^f_{\mathbf{m}\beta};$$

$$\{M_{j(\mathbf{n}\alpha)} \to M_{j(\mathbf{m}\beta)}\})|^2 W(M^f_{\mathbf{n}\alpha}) \prod_{j(\neq \mathbf{n}\alpha)} W(M_{j(\mathbf{n}\alpha)})$$

$$\cdot \delta\Big[\omega^f(M^f_{\mathbf{n}\alpha} - M^f_{\mathbf{m}\beta}) + \tilde{\omega}^0\Big(\sum_{j(\neq \mathbf{n}\alpha)} M_{j(\mathbf{n}\alpha)} - \sum_{j(\neq \mathbf{m}\beta)} M_{j(\mathbf{m}\beta)}\Big)\Big], \quad (22)$$

where $L^f_{\mathbf{n}\alpha;\mathbf{m}\beta}$ is obtained from Eq. (8), while the occupations $W(M^f_{\mathbf{n}\alpha}$ and $W(M_{j(\mathbf{n}\alpha)})$ differ from $W(M_j)$, which was given earlier, by the replacement of ω^0 by ω^f and $\tilde{\omega}^0 \equiv \mu_B g_0 \tilde{H}/\hbar$. In order to calculate (22) it is convenient to pass to the quantities

$$r = (S-1) - M_{\mathbf{n}\alpha}{}^f, \quad p = M_{\mathbf{m}\beta}{}^f - M_{\mathbf{n}\alpha}{}^f, \quad r_1 = S - M_{\mathbf{m}\beta(\mathbf{n}\alpha)},$$

$$p_1 = M_{\mathbf{n}\alpha(\mathbf{m}\beta)} - M_{\mathbf{m}\beta(\mathbf{n}\alpha)}, \quad r_j = S - M_{j(\mathbf{n}\alpha)}, \quad p_j = M_{j(\mathbf{m}\beta)} - M_{j'(\mathbf{n}\alpha)},$$

in which r, r_1, r_j are numbers of levels in the multiplets; $(0 \leqslant r \leqslant 2(S-1), \quad 0 \leqslant r_1 \leqslant 2S, \quad 0 \leqslant r_j \leqslant 2S, \quad -2(S-1) \leqslant p \leqslant 2(S-1), \quad -2S \leqslant p_1 \leqslant 2S, \quad -2S \leqslant p_j \leqslant 2S$ (the ions $j(\mathbf{m}\beta)$ and $j'(\mathbf{n}\alpha)$ with $j, j' \neq n\alpha, m\beta$ are considered equivalent relative to their PIs $\mathbf{n}\alpha$ and $\mathbf{m}\beta$). Then in place of (22) we have

$$W^f_{\mathbf{n}\alpha;\,\mathbf{m}\beta} = \frac{8\pi}{\hbar^2} |\mathscr{M}^f_{\mathbf{n}\alpha;\,\mathbf{m}\beta}|^2 \sum_{\substack{rr_1r_j \\ pp_1p_j}} \prod_{j(\neq \mathbf{n}\alpha,\ \mathbf{m}\beta)} [d^s_{S-r_j S-r_j+p_j} \cdot (\theta_{j(\mathbf{m}\beta)} - \theta_{j(\mathbf{n}\alpha)})]^2$$

$$\cdot \Big| \sum_{M_1{}^f M_2{}^f} d^{s-1}_{S-1-r\,M_1{}^f}(\theta^f_{\mathbf{n}\alpha} - \theta_{\mathbf{n}\alpha(\mathbf{m}\beta)})\, d^{s-1}_{S-1-r+p\,M_2{}^f}(\theta^f_{\mathbf{m}\beta} - \theta_{\mathbf{m}\beta(\mathbf{n}\alpha)})$$

$$\cdot \mathbf{K}_{\mathbf{n}\alpha}(M_1{}^f \to S - r_1 + p_1)\, \mathbf{K}^*_{\mathbf{m}\beta}(M_2{}^f \to S - r_1)|^2 W(r) W(r_1) W(r_j)$$

$$\cdot \delta\Big(\omega^f p + \tilde{\omega}^0 p_1 + \sum_j \tilde{\omega}^0 p_j\Big). \quad (23)$$

Here, $W(r)$, $W(r_1)$, $W(r_j)$ are the equilibrium distribution functions for the multiplet levels, which are measured from the position of the lowest energy levels of each. These functions are obtained from the expressions presented earlier by the substitutions $M^f_{\mathbf{n}\alpha} \to r$, $M_{\mathbf{m}\beta(\mathbf{n}\alpha)} \to r_1$, $M_{j(\mathbf{n}\alpha)} \to r_j$. We will assume that the exchange interaction of a PI with the neighboring ions is not too large, so that the ratio $|I^f|/ZI$ can be considered a small parameter. Then if ion $\mathbf{n}\alpha$ is excited while ion j is its nearest neighbor on the opposite sublattice (as it is for ion $\mathbf{m}\beta$), then we can assume that the inclination of the SQA

$$\Delta\theta_j = \theta_{j(\mathbf{n}\alpha)} - \theta_{j(\mathbf{m}\beta)} = \tilde{\theta}_j - \theta_j$$

is small. When the value of the parameter $|I^f|$ is smaller than I, the frequency ω^f will be smaller than the frequency $\tilde{\omega}^0$ (this relation is valid in the nearest-neighbor approximation, when ions j and $\mathbf{m}\beta$ occupy equivalent positions relative to the PI at $\mathbf{n}\alpha$). Because of the condition $\omega^f < \tilde{\omega}^0$, we can assume that the following inequality holds:

$$\exp(-\tilde{\omega}^0/k_B T) \ll \exp(-\omega^f/k_B T),$$

which in turn allows us to limit the sum in expression (23) to terms with $r_1 = r_j = 0$. The block

$$J_{pp_1} = \sum_{p_j} \prod_j [d^s_{sS+p\ j}(\Delta\theta_j)]^2 \delta\Big(\omega^f p + \tilde{\omega}^0 p_1 + \tilde{\omega}^0 \sum_j p_j\Big)$$

which arises in this case can be calculated for small deviations $\Delta\theta_j$ by the method set out in Ref. 13, giving

$$J_{pp_1} = e^{-a} \sum_m \frac{a^m}{m!} \delta(\omega^f p + \tilde{\omega}^0 p_1 - \tilde{\omega}^0 m), \quad (24)$$

where the parameter a in the magnetic Debye-Waller factor has the form

$$a = \sum_j \Big(\Big(\frac{S}{2}\Big)^{1/2} \Delta\theta_j\Big)^2 = Z\frac{S}{2}(\Delta\theta)^2. \quad (25)$$

The expression for $\Delta\theta$ is given in Ref. 13, and for the case $|I^f|/ZI \ll 1$ has the form

$$\Delta\theta \approx \frac{1}{Z}\Big[\xi^f \frac{H_E{}^f}{H_E} \sin(\theta^f + \theta) - \sin 2\theta\Big]. \quad (26)$$

Because $a < 1$, it is sufficient to retain only terms with $m = 0$ in the sum over m in Eq. (24). Finally we obtain in place of (23)

$$W^f_{n\alpha;\,m\beta} = \frac{8\pi}{\hbar^2\tilde{\omega}^0} e^{-a} |\mathcal{M}^f_{n\alpha;\,m\beta}|^2 \sum_{rp} W(r)$$
$$\cdot \Big| \sum_{M_1^f M_2^f} d^{s-1}_{S-1-r\,M_1^f}(\theta^f_{n\alpha} - \theta_{n\alpha\,(m\beta)})$$
$$\cdot d^{s-1}_{S-1-r+pM_2^f}(\theta^f_{m\beta} - \theta_{m\beta\,(n\alpha)})$$
$$\cdot \mathbf{K}_{n\alpha}\left(S - \frac{\omega^f}{\tilde{\omega}^0} p \dashrightarrow M_1^f\right) \mathbf{K}^*_{m\beta}(S \to M_2^f)\Big|^2. \quad (27)$$

Expression (27) is valid when the condition $\omega^f p + \tilde{\omega}^0 p_1 = 0$ is fulfilled, which is the value contained in the δ-function in (24) for $m = 0$. If, however, the frequencies ω^f and $\tilde{\omega}^0$ are such that this condition cannot hold for attainable values of p and p_1, then only the case $p = 0$ is possible (and correspondingly $p_1 = 0$). Let us write down the probability of a jump for a self-trapped exciton in this case, setting $p = 0$ in (27) and taking into account the fact that according to (7) and the figure,

$$\mathbf{K}_{n\alpha}(M_1^f \dashrightarrow S)\mathbf{K}^*_{m\beta}(M_2^f \dashrightarrow S)$$
$$= \frac{1}{2}\delta_{M_1^f, M_2^f}\delta_{M_1^f, S-1}\cos^2\left(\frac{\theta_{n\alpha\,(m\beta)} - \theta_{m\beta\,(n\alpha)}}{2}\right).$$

Therefore, taking into account the relations given in (10), we have

$$W^f_{n\alpha;m\beta} = \frac{2\pi}{\hbar^2\tilde{\omega}^0} e^{-a} |\mathcal{M}^f_{n\alpha;m\beta}|^2 \sum_r W(r)\,[d^{s-1}_{S-1-rS-1}(\theta^f - \tilde{\theta})]^2$$
$$\cdot \left[\delta_{\alpha\beta}\cos^4\frac{\tilde{\theta}-\theta}{2} + (1-\delta_{\alpha\beta})\cos^4\tilde{\theta}\right]. \quad (28)$$

In the nearest-neighbor approximation, the frequency $\tilde{\omega}^0$ practically coincides with ω^0 for small values of $|I^f|ZI$ in the interval of fields $2H_E\sigma \geqslant H_0 \geqslant H_{er}$. Let the temperature be close to zero, so that we can limit ourselves only to terms with $r = 0$ in the sum. For simplicity, we will include the variation in SQA only for the PI (besides the magnetic Debye-Waller factor). Then from (28) we obtain

$$W^f_{n\alpha;m\beta} = \frac{2\pi}{\hbar^2\omega^0} e^{-a}\cos^{8(s-1)}\left(\frac{\theta^f - \theta}{2}\right)[\,|\mathcal{M}_1^f|^2\delta_{\alpha\beta} + \cos^4\theta\,|\mathcal{M}_2^f|^2$$
$$\cdot(1-\delta_{\alpha\beta})\,]. \quad (29)$$

As is clear from (25), (26), when the value of I^f is not too large, we can set

$$a \approx \frac{S}{2Z}\sin^2 2\theta = \frac{2S}{Z}\left(\frac{M}{M_0}\right)^2\left[1 - \left(\frac{M}{M_0}\right)^2\right]. \quad (30)$$

In Eq. (30) we have introduced the ratio of M and M_0, where M is the magnetization induced in the crystal by the magnetic field and M_0 is the saturation magnetization, in the form $\cos\theta = M/M_0$. We also write the hopping probabilities (29) for the self-trapped exciton for the ion's own sublattice $[W_1^f(M)]$ of the AFI. For simplicity we will investigate the case $I^f \approx 0$. Then, according to (21), we have $\theta^f = 0$, and consequently,

$$W_1^f(M) = \frac{2\pi}{\hbar\mu_B g_0 H_E} e^{-a}|\mathcal{M}_1^f|^2\left[\frac{1}{2}\left(1 + \frac{M}{M_0}\right)\right]^{4(s-1)}, \quad (31)$$

$$W^f(M) = \frac{2\pi}{\hbar\mu_B g_0 H_E} e^{-a}|\mathcal{M}^f|^2\left(\frac{M}{M_0}\right)^4\left[\frac{1}{2}\left(1 + \frac{M}{M_0}\right)\right]^{4(S-1)}. \quad (32)$$

The exciton diffusion coefficients in the direction along the ions of its own sublattice and along the direction corresponding to those ions which belong to the nearest neighbors on the opposite sublattice will be proportional to the quantities (31) and (32), respectively. In the field interval $H_{cr} \leqslant H_0 \leqslant 2H_E$ the quantity M varies over the range $0 \lesssim M \leqslant M_0$. In this case the factor $\exp(-a)$ varies slightly and this variation can be neglected for estimation purposes. Setting $S = 5/2$ (for an AFI containing Mn^{2+} ions), we see that according to (31) the diffusion coefficient increases by a factor of 2^6 for diffusion along the ions of its own sublattice because the magnetization increases from 0 to $M = M_0$, depending on whether or not the SQA of the PI is unchanged. As for diffusion along the nearest neighbors from the opposite sublattice, in Ref. 10 it was observed that the diffusion coefficient varied as $(M/M_0)^4$ in the field interval up to 70 kOe, in keeping with (32). The presence of an additional factor of $[(1/2)(1 + M/M_0)]^{4(s-1)}$ is revealed at higher fields, if $I^f \approx 0$. If, however, $I^f \neq 0$, then for $|I^f|/ZI < 1$, we can make use of the more general Eq. (29), taking (31) into account. If $I^f \neq 0$ and $|I^f| > ZI$, then a more detailed calculation is required for the imhomogeneous magnetization which forms around the PI[13].

The analysis we have carried out here shows that the variation of the SQAs of a PI and of its neighboring ions leads to a decrease in the magnitude of the resonance interaction between magnetic ions and to a transformation of free excitons into self-trapped excitons. For weak relaxation within the multiplets, this contributes to a decrease in the exciton band parameters (which is larger the larger the spin cloud around the PI and to complicated dependences of these parameters on the external magnetic field. For strong relaxation within the multiplet, an equilibrium spin cloud forms near the PI, which moves along with the PI as a self-trapped exciton; this incoherent motion appears as diffusion. The diffusion coefficient depends significantly on the direction of propagation of the self-trapped exciton and the character of the spin noncollinearity.

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[1)] The SQA is chosen so that for weak single-ion anisotropy the projection of the spin on the SQA is always a good quantum number for the magnetic ion.

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