Striction effects and dynamics of the magnetic subsystem in spin-reorientation phase transitions. Symmetry aspects

V. G. Bar'yakhtar, I. M. Vitebskiĭ, Yu. G. Pashkevich, V. L. Sobolev, and V. V. Tarasenko

Physicotechnical Institute, Academy of Sciences of the Ukrainian SSR, Donetsk

Institute of Theoretical Physics, Academy of Sciences of the Ukrainian SSR, Kiev

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A study is made of the dynamics of a spin system in the vicinity of a spin-reorientation phase transition of the soft mode type. A criterion of the appearance of a striction gap in the oscillation spectrum of the order parameter is formulated and the nature of this gap is identified. It is shown that a striction gap of purely exchange origin may exist.

The majority of the know spin-reorientation phase transitions are of the soft mode type when considered from the dynamic point of view. This means that the coresponding frequency of the normal oscillations of the spin system in the vicinity of the critical point may be anomalously low. Nevertheless, at the critical point itself the frequency of the normal mode $\omega = \omega(T)$ which becomes softer may remain finite because of the striction effects. Then, a singularity appears in the vibration spectrum of the elastic subsystem: the dispersion law of sound changes from linear to quadratic.

It follows from this brief introduction that the magnetoelastic interactions play the dominant role in the formation of a gap in the spectrum of magnetic oscillations at the spin-reorientation phase transition (SRPT) point. The history of this topic can be found in a review of Turov and Shavrov,[1] where the essential references are cited. Numerous theoretical investigations of the topic have been concerned only with special simplest types of the SRPT in ferromagnets and antiferromagnets (see, for example, Refs. 2–5). Detailed calculations have been carried out for such cases and whenever possible a comparison has been made with the available experimental results. However, the general symmetry aspects of the topic have been ignored. The following problems still remain.

1. A striction gap in the oscillation spectrum of the magnetic order parameter appears at the critical point of all the SRPTs investigated so far (we are speaking here of theoretical treatments). The only exception are those cases in which the absence of a gap is a consequence of model approximations. A natural question thus arises: how common is such behavior? Moreover, if the reverse is possible, then what is the general criterion of the existence of a striction gap in the spectrum of magnetic excitations corresponding to a soft mode?

2. It has been assumed so far that the striction gap is due to relativistic interactions and, in particular, due to the relativistic magnetostriction. In the earlier investigations this has indeed been true: the adoption of the exchange approximation has always destroyed the gap in the soft-mode spectrum. Nevertheless, it is found that, in principle, there may be situations in which the striction gap in the oscillation spectrum of the magnetic order parameter is entirely due to the exchange interaction. Such situations can be identified without carrying actual calculations but applying only pure symmetry considerations.

These problems are the subject of the present paper. We shall describe a phase transition using the mean-field theory. The role of fluctuations in problems of interest to us is clearly of very minor importance. This has been pointed out repeatedly by many authors in connection with studies of orientational phase transitions in magnetic materials.[1] The effects associated with the finite size of real crystals and with the damping will not be considered.

1. STRICTION GAP IN THE SPECTRUM OF A SOFT MODE CONSIDERED ON THE BASIS OF THE LANDAU THEORY

The appearance of a striction gap in the oscillation spectrum of an order parameter can with advantage be demonstrated on the basis of the phenomenological theory of Landau. We shall consider a second-order phase transition which is classified on the basis of symmetry as a proper ferroelastic transition. We recall that in this case the Landau expansion for the free energy W contains an invariant which is linear in respect of the order parameter and of strains:

$$W={}^1/_2 a\eta^2+{}^1/_4 b\eta^4-\lambda\eta u+{}^1/_2 Ku^2. \tag{1}$$

Here, η is the order parameter of the phase transition and u is a certain combination of the components of the strain tensor $u_{\alpha\beta}$. We shall use the simplified symbolic notation. In general, η and u are multicomponent quantities (if the phase transition occurs in accordance with a multidimensional irreducible representation of the symmetry group of the high-symmetry phase).

The equilibrium values of η and u can be found from a minimum of Eq. (1) describing W:

$$\bar{\eta}=\begin{cases} 0, & a^*\geqslant 0 \\ (-a^*/b)^{1/2}, & a^*\leqslant 0 \end{cases}, \tag{2}$$

$$\bar{u}=\lambda\bar{\eta}K^{-1}, \tag{3}$$

where

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$$a^{*}=a-\lambda^{2}K^{-1}. \tag{4}$$

The phase transition point corresponds to $a^{*}=0$.

In the absence of damping the frequency of oscillations of the order parameter is governed by the generalized "stiffness" of the system:

$$\varkappa^{-1}=(\partial^{2}W/\partial\eta^{2})_{u=\bar{u}}. \tag{5}$$

Equation (5) represents the frozen-lattice concept. In the case of an infinite crystal it is found that homogeneous strains cannot vary at a finite rate (i.e., $u_{\alpha\beta}$ is independent of time) and, therefore, they cannot follow oscillations of the order parameter η. Therefore, in studies of homogeneous oscillations in an unbounded sample the crystal lattice should be regarded as static, i.e., as "frozen" (Ref. 1).

Using Eqs. (1)–(5), we obtain

$$\varkappa^{-1}=a+3b\bar{\eta}^{2}=\begin{cases} a^{*}+\lambda^{2}K^{-1}, & a^{*}\geqslant 0 \\ 2|a^{*}|+\lambda^{2}K^{-1}, & a^{*}\leqslant 0 \end{cases}. \tag{6}$$

It should be stressed that $\varkappa^{-1}$ is always greater than the usual static generalized reciprocal susceptibility χ^{-1}, which is given by the expression

$$\chi^{-1}=\frac{d^{2}}{d\eta^{2}}W(\eta,u(\eta))=a^{*}+3b\eta^{2}=\begin{cases} a^{*}, & a^{*}\geqslant 0 \\ 2|a^{*}|, & a^{*}\leqslant 0 \end{cases}. \tag{7}$$

It should be noted that $u(\eta)$ in Eq. (7) is equal to $\lambda\eta K^{-1}$, which is a consequence of static adjustment of the lattice so that the order parameter has its equilibrium value [see Eq. (3)].

It follows from Eq. (6) and Fig. 1 that $\varkappa^{-1}$ is rigorously positive throughout the temperature range under discussion, including the critical point, whereas χ^{-1} of Eq. (7) vanishes at the phase transition point.

The temperature dependence of the oscillation frequency of the order parameter is similar to the dependence $\varkappa^{-1}(T)$. Model calculations indicate that usually (but not always!) we have

$$\omega^{2}(T)\propto\varkappa^{-1}(T). \tag{8}$$

This problem is discussed in greater detail in Ref. 6. The striction gap at the critical point itself (i.e., at $a^{*}=0$) is governed by the corresponding value of $\varkappa^{-1}$, i.e., it is governed by the quantity $\lambda^{2}K^{-1}$.

The above discussion demonstrates clearly the inevitability of the appearance of a striction gap in the oscillation spectrum of the order parameter at the point of a proper ferroelastic phase transition, i.e., when the expansion of the thermodynamic potential contains invariants of the ηu type. We can easily show that the invariants with higher powers of η or u do not contribute to the formation of a gap in the soft-mode spectrum at the critical point.

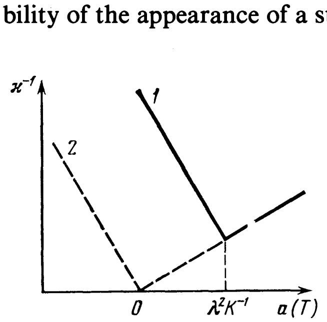

FIG. 1. Temperature dependence of the generalized dynamic stiffness in the vicinity of the critical point: 1) $\lambda\neq 0$; 2) $\lambda=0$.

The invariants of the ηu type are known to be absent if a phase transition is accompanied by "multiplication" of the primitive (magnetic or crystallographic) cell or by the loss of a center of inversion (corresponding to the $\bar{1}$ or $\bar{1}'$ operations of the magnetic symmetry group) or if the phase transition does not alter the crystallographic system, for example, if before and after the transition a crystal is cubic or tetragonal, etc.; in the latter case a spontaneous striction always corresponds to a single representation of the symmetry group of the high-symmetry phase and, consequently, such a phase transition is in no way ferroelastic.

We shall now consider SRPTs from the viewpoint presented above. In the simplest case an SRPT reduces to rotation of the magnetic structure as a whole in the spin space without a significant change of the angles between the atomic spins. If the magnetic structure is collinear (even if only in the exchange approximation), which may mean that the structure is ferromagnetic, antiferromagnetic, or ferrimagnetic, then an SRPT in such a system is always a proper ferroelastic transition and a striction gap in the soft-mode spectrum is of the relativistic origin.[1)] These are the cases that have been discussed so far in the literature and there is no point in dealing with them in greater detail.

However, if the magnetic structure of a crystal is strongly noncollinear, the situation may be different. By way of example, we can mention here a spin reorientation in Mn_3NiN in which the magnetic structure before, during, and after completion of the reorientation process is described by magnetic groups which belong to the rhombohedral symmetry.[2)] Consequently, both phase transitions corresponding to the onset and completion of the spin reorientation process are not ferroelastic and there is no striction gap in the spectrum of spin excitations at either of the critical points.

An example of an SRPT which does not reduce to rotation of a magnetic structure as a whole in the spin space is the collapse of magnetic sublattices in an antiferromagnet subjected to a strong magnetic field.

If a sufficiently strong external magnetic field (exceeding the exchange interaction field) is applied to a crystal, then any magnetic structure becomes ferromagnetic, i.e., all the magnetic sublattices become oriented parallel to the external magnetic field. A phase transition associated with vanishing of the angle between the sublattice magnetizations is known as the collapse of sublattices. Therefore, the variety of the sublattice collapse transitions is as great as of the magnetic structures in general.

In a case of this kind it is not possible to give a unique answer to the question of whether a striction gap can exist in the spectrum of magnetic excitations at the critical point. The only general conclusion is that if a striction gap does appear at the sublattice collapse point, it is of the relativistic origin, i.e., it disappears in the exchange approximation. We shall prove this conclusion in the next section and concentrate here in greater detail on a special case which is a sublattice collapse transition in two-sublattice antiferromagnets.

If an external magnetic field **H** is greater than the collapse transition field H_e, then the magnetic structure of the antiferromagnet in question becomes transformed into a collinear ferromagnetic structure. We shall assume that $\mathbf{H}\|z$, so that if $H>H_e$, we have

$$\mathbf{m}\|\mathbf{H}\|z, \quad \mathbf{l}=0, \tag{9}$$

where **l** is the antiferromagnetic vector and **m** is the total magnetic moment of the sublattices.

If $H<H_e$, then $\mathbf{l}\neq 0$ and the magnetic structure becomes noncollinear. The order parameter for the phase transition in a field $H=H_e$ is one of the transverse components of the antiferromagnetic vector, for example, l_y (if the z axis coincides with the crystallographic axis of symmetry higher than twofold, then the quantities l_x and l_y are the components of the same two-component order parameter). The fullest symmetry analysis of sublattice collapse transitions in two-sublattice antiferromagnets of different crystallographic symmetries can be found in Ref. 9. Numerous specific examples are also considered there.

It follows from the general criterion presented above that a striction gap appears in the spectrum of magnetic excitations at $H=H_e$ if and only if one of the transverse components of the antiferromagnetic vector **l** and one of the components of the strain tensor $u_{\alpha\beta}$ can be combined to form a product lu, which is invariant relative to the magnetic symmetry group G_{fm} of the high-field ferromagnetic phase described by Eq. (9).[3)] This in its turn is possible only if there are invariants of the mlu type relative to the symmetry group of the paramagnetic phase G_{pm}, i.e., in the presence of a linear piezomagnetic effect in the antiferromagnetic state. We shall not consider all possible situations which may arise for different types of crystallomagnetic structures, because the analysis is essentially trivial and it is more reasonable to carry it out separately for each specific case. Here, we shall consider a unique case of orthorhombic two-sublattice antiferromagnets.

Clearly, a sublattice collapse transition may be a proper ferroelastic transition only if the crystallochemical primitive cell is identical with the magnetic cell in the antiferromagnetic state. Therefore, the analysis may be limited to the case of magnetic ordering characterized by $\mathbf{k}=0$ using the point symmetry group of a crystal.

The standard classification of the components of the vectors **l** and **m** and also of the components of the strain tensor $u_{\alpha\beta}$ in accordance with the irreducible representations of the point symmetry group of the paramagnetic phase D_{2h} (Ref. 6) shows that two types of magnetic ordering are possible in an orthorhombic crystal. Type I ordering admits the existence of weak ferromagnetism, i.e., it admits the possibility of the Dzyaloshinskii interaction, whereas type II ordering does not admit this interaction. The existence of invariants of the mlu type is possible only in type I antiferromagnets. Consequently, in type II orthorhombic antiferromagnets, as well as in antiferromagnets with different magnetic and crystallochemical cells, a sublattice collapse transition is not accompanied by the appearance of a striction gap in the magnon spectrum.

In the case of orthorhombic type I antiferromagnets (which include in particular orthoferrites[8]) a sublattice collapse transition is possible only when the magnetic field **H** is oriented along one of the crystallographic axes. Only in this case is a sublattice collapse transition a proper ferroelastic transition and a striction gap of the relativistic origin appears in the antiferromagnetic resonance spectrum at $H=H_e$. A calculation carried out in the usual way (for details see Ref. 6) gives the following expression for the striction gap Δ in the spectrum of magnetic excitations at the sublattice collapse point:

$$\Delta\propto(H_A\lambda^2K^{-1})^{1/3}, \tag{10}$$

where H_A is the orthorhombic anisotropy field. It should be noted that the expression for the gap Δ does not include the exchange field. Therefore, the exchange enhancement of the magnetoelastic interaction (for details see Ref. 1) does not occur in this case.

2. POSSIBILITY OF EXISTENCE OF A STRICTION GAP OF EXCHANGE ORIGIN IN THE SPECTRUM OF A SOFT MODE AT A SPIN-REORIENTATION PHASE TRANSITION

The necessary infomation from the theory of the exchange symmetry can be found in Refs. 10–14. Whenever possible, we shall use the terminology adopted in these papers.

In the exchange approximation the spectrum of magnetic excitations of a magnetically ordered crystal must necessarily include activation-free Goldstone modes,[4)] which are usually called acoustic magnons. Activation occurs only if we allow for the relativistic interactions, particularly for the relativistic magnetostriction. In this connection it is necessary to specify which striction gap in a spectrum we are speaking of if the exchange approximation is adopted. This is necessary because there is an extensive class of SRPTs involving considerable changes of the angles between the atomic spins. They include also the sublattice collapse transition discussed above. Normal oscillations of the spin system corresponding to a mode which becomes softer can then have a very different symmetry which is not the symmetry of acoustic magnons with the same wave vector (in the present case we shall be interested in homogeneous oscillations, i.e., we shall consider only the modes with $\mathbf{k}=0$). In other words, an optical (exchange) mode may become softer, whereas acoustic oscillations of the spin symmetry do not participate in the phase transition and do not even interact with the soft mode. If in addition it is found that such an SRPT is a proper ferroelastic transition even in the exchange approximation, then the frequency of oscillations of the order parameter remains finite at the critical point and the value of this frequency is governed entirely by the exchange interactions, particularly by the exchange magnetostriction.

Neither a sublattice collapse transition nor phase transitions of the kind that occur in ferrites in strong magnetic fields[5] are proper ferroelastic transitions in the exchange approximation. This means that even if the parameter λ of Eq. (4) differs from zero for these cass, this occurs only because of the relativistic interactions. This represents a special case of the following general conclusion: if at the critical point of an SRPT the magnetic structure is collinear (at least in the exchange approximation), then in the exchange approximation such a phase transition is not a proper ferroelastic transition. In fact, we shall assume that at the critical point (and, therefore, in the high-symmetry phase) the magnetic structure is collinear. Then, the atomic spins are either parallel or antiparallel to, for example, the z axis. The order parameter governing the disymmetric phase at an SRPT is a certain linear combination of the x and y components of the atomic spins.[5)] Clearly, rotation in the spin space around the z axis by an angle π reverses always the sign of the order parameter. However, the exchange energy and the exchange striction are not affected, in accordance with the definition of the exchange approximation. Consequently, the exchange striction cannot in this case be linear in respect of the order parameter of an SRPT. The transition itself is not a proper ferroelastic transition in the exchange approximation.

We shall make *in passim* an important remark not related directly to the above discussion. We shall consider an arbitrary collinear (in the exchange approximation) magnetic structure oriented specifically along the z axis. Let $\psi(\mathbf{r})$ be an arbitrary linear combination of the x and y components of the atomic spins, which depends in any manner on the coordinates (for example, it may represent a magnon with an arbitrary wave vector). Following exactly the analysis made above, we can easily show that the exchange approximation forbids any interactions linear in $\psi(\mathbf{r})$ and of the type

$$\psi(\mathbf{r})\,p(\mathbf{r}),$$

where $p(\mathbf{r})$ describes any "nonspin" physical quantity such as an atomic displacement (phonon), electric dipole moment, etc. Hence, it follows that, in particular, processes such as one-magnon absorption of the energy from an alternating electric field and a magnetoacoustic resonance are possible in the exchange approximation only in noncollinear magnetic structures.

Therefore, a striction gap in the spectrum of a soft mode is entirely of the exchange origin if an SRPT is a proper ferroelastic transition already in the exchange approximation. This is possible if an SRPT occurs between two noncollinear (in the exchange approximation) magnetic structures. In a formal group-theoretic analysis we must bear in mind that a satisfactory description of the exchange symmetry is given by color symmetry groups of the P or Q type. Consequently, the magnetic order parameter of an SRPT should correspond to an irreducible representation of the color symmetry group of the high-symmetry phase. However, we shall not consider the formal aspects of this problem and discuss instead a specific example of an exchange-striction gap in the spectrum of a soft mode.

Figure 2a shows one of the possible types of magnetic ordering in a tetragonal lattice. We shall consider a spin reorientation in which the sublattice magnetizations rotate as shown in Fig. 2b and thus form a collinear antiferromagnetic structure (phase c, shown in Fig. 2c).

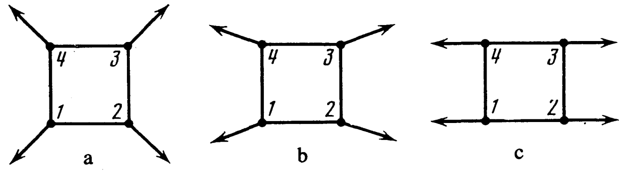

FIG. 2. Spin reorientation in a tetragonal lattice. The magnetic moments lie in the basal plane xy.

The calculation reported below shows that both phase transitions corresponding to the onset (Fig. 2a) and completion (Fig. 2c) of a spin reorientation are accompanied by softening of the optical (exchange) mode of homogeneous oscillations of the spin subsystem. Without allowance for the magnetostriction the revelant frequencies vanish at the critical points. An allowance for the exchange striction gives rise to a gap (of the exchange origin) in the spectrum of oscillations of the order parameter only at the first of the critical point (corresponding to the phase $a \rightleftarrows$ phase b transition). In the case of the phase $b \rightleftarrows$ phase c transition a gap does not appear even if we allow for the relativistic interactions because this phase transition is accompanied by a change in the translation symmetry and cannot be a proper ferroelastic transition.

The acoustic oscillations of the spin system do not participate in phase transitions in the situation shown in Fig. 2, i.e., they do not even interact with a soft mode because they have a different symmetry.

We shall now calculate the frequencies of homogeneous oscillations of the spin system for the magnetic structures shown in Fig. 2. The Hamiltonian of the quadratic exchange interaction is

$$\hat{\mathscr{H}}_e^{(2)} = J_A(\hat{\mathbf{A}}^2+\hat{\mathbf{B}}^2)+J_C\hat{\mathbf{C}}^2+J_F\hat{\mathbf{F}}^2, \tag{11}$$

where

$$\begin{aligned} &\hat{\mathbf{A}}=\hat{\mathbf{S}}_1-\hat{\mathbf{S}}_2-\hat{\mathbf{S}}_3+\hat{\mathbf{S}}_4, \quad \hat{\mathbf{C}}=\hat{\mathbf{S}}_1-\hat{\mathbf{S}}_2+\hat{\mathbf{S}}_3-\hat{\mathbf{S}}_4,\\ &\hat{\mathbf{B}}=\hat{\mathbf{S}}_1+\hat{\mathbf{S}}_2-\hat{\mathbf{S}}_3-\hat{\mathbf{S}}_4, \quad \hat{\mathbf{F}}=\hat{\mathbf{S}}_1+\hat{\mathbf{S}}_2+\hat{\mathbf{S}}_3+\hat{\mathbf{S}}_4. \end{aligned} \tag{12}$$

If the exchange constants in Eq. (11) satisfy the condition

$$J_A<J_C,\ J_F, \tag{13}$$

then the ground state of the Hamiltonian of Eq. (11) is

$$\mathbf{A}\neq 0,\quad \mathbf{B}\neq 0,\quad \mathbf{F}=\mathbf{C}=0, \tag{14}$$

where

$\mathbf{A}=\langle\hat{\mathbf{A}}\rangle,\quad \mathbf{B}=\langle\hat{\mathbf{B}}\rangle,\quad \mathbf{C}=\langle\hat{\mathbf{C}}\rangle,\quad \mathbf{F}=\langle\hat{\mathbf{F}}\rangle.$

If temperatures are sufficiently low, we can assume that

$$S_1{}^2=S_2{}^2=S_3{}^2=S_4{}^2\equiv S^2. \tag{15}$$

Allowing for Eq. (14), we find that the normalization conditions of Eq. (15) become

$$\mathbf{A}^2+\mathbf{B}^2=(4S)^2,\quad (\mathbf{A},\mathbf{B})=0,\quad \mathbf{F}=\mathbf{C}=0. \tag{16}$$

Finally, assuming that $A\parallel x$ and $B\parallel y$ [such a selection is permissible within the framework of the exchange approximation allowing for Eq. (16)], we find that the magnetic structures shown in Fig. 2 are obtained.

It should be pointed out that in the case defined by Eq. (13) the quadratic exchange interaction of Eq. (11) does not even fix the relative orientations of the atomic spins and that the energy of the quadratic exchange is the same for all three structures shown in Fig. 2. We can determine uniquely the ground state within the exchange approximation framework if we supplement the Hamiltonian of Eq. (11) with the exchange terms of higher orders in respect of the spin:

$$\hat{\mathscr{H}}_e^{(4)}=P(\hat{\mathbf{A}}^2-\hat{\mathbf{B}}^2)^2,\quad \hat{\mathscr{H}}_e^{(8)}=Q(\hat{\mathbf{A}}^2-\hat{\mathbf{B}}^2)^4. \tag{17}$$

The exchange in energy deduced for the AB-type ordering from Eqs. (11), (16) and (17) is

$$W_e\equiv\langle\mathscr{H}_e\rangle=J_A(\mathbf{A}^2+\mathbf{B}^2)+P(\mathbf{A}^2-\mathbf{B}^2)^2+Q(\mathbf{A}^2-\mathbf{B}^2)^4. \tag{18}$$

The SRPT shown in Fig. 2 occurs because of a change in the sign of the biquadratic exchange parameter P. The ground state of a magnetic system is found from the condition for a minimum of the energy W_e described by Eq. (18) and, as a function of P, it has the following value for the three phases:

1) phase a:

$$A=B=4S/\sqrt{2}, \tag{19}$$

which is stable in the range $P>0$;

2) phase b:

$$A^2-B^2=(-P/2Q)^{1/4}, \tag{20}$$

which is stable in the range $-2Q(4S)^4\leqslant P\leqslant 0$;

3) phase c:

$$A=4S,\quad B=0, \tag{21}$$

which is stable in the range $P<-2Q(4S)^4$.

A calculation of the frequencies of homogeneous oscillations of the spin system will be carried out using a scheme described in Ref. 15, i.e., we shall begin from quantum equations of motion for symmetrized operators of Eq. (12):

$$i\hbar\dot{\hat{\Gamma}}_\alpha=[\hat{\Gamma}_\alpha,\hat{\mathscr{H}}_e],\quad \alpha=x,y,z, \tag{22}$$

where

$$\hat{\mathscr{H}}_e=\hat{\mathscr{H}}_e^{(2)}+\hat{\mathscr{H}}_e^{(4)}+\hat{\mathscr{H}}_e^{(8)}, \tag{23}$$

and $\hat{\Gamma}_\alpha$ is one of the twelve symmetrized operators of Eq. (12). After linearization of these equations in the random phase approximation and subject to Eqs. (19)–(21), we obtain the following expressions for the frequency of a soft optical mode of each of the three magnetic configurations of Fig. 2:

phase a:

$$\omega=64S^2[P(J_C-J_A)]^{1/2}; \tag{24}$$

phase b:

$$\omega=64S^2\left[-2P\left(1+\frac{P}{2(4S)^4Q}\right)(J_C-J_A)\right]^{1/2}; \tag{25}$$

phase c:

$$\omega=64S^2[-(P+2Q(4S)^4)(J_C-J_A)]^{1/2}. \tag{26}$$

Therefore, without allowance for the magnetostriction the soft-mode frequency vanishes at both critical points.

In addition to the soft optical mode of Eqs. (24)–(26), there are also three acoustic magnon modes in the phases a and b, and only two modes in the collinear phase c. If $\mathbf{k}=0$ their frequencies differ from zero only in the presence of the relativistic interactions. There is not much point in giving the relevant expressions. We shall simply point out that in the situation shown in Fig. 2 the acoustic modes do not interact with a soft mode when $\mathbf{k}=0$ because they have a different symmetry.

Clearly, in the phases a and b a soft mode corresponds to homogeneous oscillations of the spin system, i.e., the oscillations with $\mathbf{k}=0$, whereas in the collinear phase c a soft mode corresponds to an irreducible representation of the magnetic symmetry group of the ground state with $\mathbf{k}\neq 0$ (a magnetic cell of the phase c is half that of the phases a and b).

We shall now consider the effect of magnetostriction. In the exchange approximation the nonisomorphous part of the magnetoelastic interaction[6] is described by the Hamiltonian

$$\hat{\mathscr{H}}_e^{(\mathrm{MY})}=-\Lambda(\hat{\mathbf{A}}^2-\hat{\mathbf{B}}^2)(u_{xx}-u_{yy})+\frac{K}{2}(u_{xx}-u_{yy})^2, \tag{27}$$

where K is the relevant elastic modulus and Λ is one of the exchange magnetostriction constants. The equilibrium deformation is given by the expression

$$\bar{u}_{xx}-\bar{u}_{yy}=\Lambda(\mathbf{A}^2-\mathbf{B}^2)K^{-1}. \tag{28}$$

Allowing for Eqs. (27) and (28), we now obtain the following magnetostriction (magnetoelastic, denoted by the index "me") correction to the ground-state energy:

$$W_e^{(\mathrm{MY})}=-\frac{\Lambda^2}{2K}(\mathbf{A}^2-\mathbf{B}^2)^2. \tag{29}$$

Comparing Eqs. (29) and (18), we can see that an allowance for the contribution of the nonisomorphous part of the exchange striction to the ground-state energy is equivalent to renormalization of the biquadratic exchange parameter $P\to P^*$, where

$$P^*=P-\Lambda^2(2K)^{-1}. \tag{30}$$

The corresponding replacement should be carried out in Eqs. (19)–(21), which give the equilibrium magnetic configurations and the ranges of their stability. As a result, we obtain

phase a:

$$A=B=4S/\sqrt{2}, \tag{31}$$

stable in the range $P^*\geqslant 0$;

phase b:

$$A^2-B^2=(-P^*/2Q)^{1/4}, \tag{32}$$

stable in the range $-2Q(4S)^4\leqslant P^*\leqslant 0$;

phase c:

$$A=4S,\quad B=0, \tag{33}$$

stable in the range $P^*\leqslant -2Q(4S)^4$.

In calculating the striction correction to the frequencies of homogeneous oscillations of the spin system we have to use again the concept of a frozen lattice. In the present case this means that the quantity $u_{xx} - u_{yy}$ in Eq. (27) should be replaced by its equilibrium value given by Eq. (28). This then gives the following correction to the Hamiltonian of Eq. (23):

$$\hat{\mathscr{H}}_e^{(MY)} = -\frac{\Lambda^2}{K}(\mathbf{A}^2-\mathbf{B}^2)(\hat{\mathbf{A}}^2-\hat{\mathbf{B}}^2)+\frac{\Lambda^2}{2K}(\mathbf{A}^2-\mathbf{B}^2)^2. \tag{34}$$

When an allowance is made for this correction, the expressions for the frequency of a soft optical mode become as follows:

phase a:

$$\omega=64S^2\left[\left(P^*+\frac{\Lambda^2}{2K}\right)(J_C-J_A)\right]^{1/2}; \tag{35}$$

phase b:

$$\omega=64S^2\left[-2\left(P^*-\frac{\Lambda^2}{2K}\right)\left(1+\frac{P^*}{2(4S)^4Q}\right)(J_C-J_A)\right]^{1/2}; \tag{36}$$

phase c:

$$\omega=64S^2[-(P^*+2(4S^4)Q)(J_C-J_A)]^{1/2}. \tag{37}$$

Therefore, an allowance for the exchange striction gives rise to activation in the spectrum of a soft optical mode at the critical point of the phase $a\rightleftarrows$ phase b transition. The striction gap is

$$\omega(P^*=0)=64S^2\left[\frac{\Lambda^2}{2K}(J_C-J_A)\right]^{1/2}. \tag{38}$$

It should be pointed out that the gap size is governed only by the parameters of the quadratic exchange Hamiltonian subject to an allowance for the quadratic (in respect of the spins) exchange striction.

In the case of the phase $b\rightleftarrows$ phase c transition with $P^* = -2(4S)^4Q$ an allowance for striction does not give rise to activation in the soft mode spectrum because this transition is not a proper ferroelastic phase transition.

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1) The exact meaning of this statement is as follows: in the exchange approximation, i.e., in the complete absence of the relativistic interactions, there is no striction gap in the soft mode spectrum.

2) More detailed information on the magnetic structure and magnetic symmetry of this crystal can be found in Ref. 7 or in the handbook of Oles *et al.*[8]

3) The components l_x and l_y themselves and any linear combination of these components cannot be invariant relative to G_{fm}. In the opposite case the phase described by Eq. (9) does not appear (strictly speaking) for the H no matter how high and the critical point of the spin reorientation transition is absent.[9]

4) Generaly speaking this is true only in the absence of an external magnetic field. However, this circumstance is unimportant in our analysis.

5) In principle, the phase transition may alter not the directions but only the absolute values of the magnetic moments of the various sublattices. However, it is clear that such a transition is not of the soft mode type. Moreover, it is not even a spin-reorientation transition.

6) This means the part that disturbs the tetragonality of the original lattice (we are speaking here of orthorhombic distortions).

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