Magnetoelastic effects in noncollinear antiferromagnets

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The static and dynamic manifestations of magnetoelastic coupling in the noncollinear antiferromagnet UO_2 are investigated theoretically. It is shown that only optical magnons interact effectively with sound, whereas all three acoustic magnon modes are weakly coupled to the elastic subsystem. The existence of a linear piezomagnetic effect in this compound is predicted. A method of calculating the magnon spectrum in the vicinity of symmetric Brillouin-zone points is demonstrated. The method takes full account of the crystallographic and magnetic symmetry of the system.

INTRODUCTION

Coupled electromagnetic waves have been the subject of many studies. The history of this topic is reported in considerable detail in Refs. 1 and 2. Almost all the available papers (with the rare exception, e.g., of Refs. 3–5) dealt only with collinear or almost-collinear magnetic structures whose noncollinearity is due to weak relativistic interactions. All the effects possible for these cases have been calculated in detail and analyzed. The magnetoelastic effects in antiferromagnets that are noncollinear even in the exchange approximation (these include UO_2, whose magnetic structure is shown in Fig. 1), however, have hardly been studied. This holds in particular for the dynamic aspects of the problem.

Our investigations have shown that the character and dynamic manifestation of the magnetoelastic coupling in UO_2 differ in principle from those previously known from the analysis mainly of collinear and weakly noncollinear ferro- and antiferromagnets.

The magnon spectrum of multisublattice magnets can contain, besides acoustic modes, also exchange (optical) modes whose activation is of purely exchange origin. If the magnetic structure is collinear (albeit in the exchange approximation), only acoustic magnons interact effectively with the sound[6] (of course, all these exchange interactions are much stronger than the relativistic ones), and this interaction is relativistic and vanishes in the exchange approximation.

The situation in UO_2 is the exact opposite. The acoustic magnons (which exist in three modes in this case) hardly interact with the sound in the region of small wave vectors. At the same time, the exchange magnon mode is strongly coupled with the elastic subsystem, with the coupling determined by exchange magnetostriction that is quadratic in the spins. This offers, in particular, a rare opportunity of exciting optical magnons by oscillations of the elastic subsystem.

MAGNETIC ORDERING IN UO_2

The UO_2 crystal symmetry is described by the $Fm3m$ Fedorov group. The uranium ions occupy a-type positions that coincide with the fcc lattice sites. The magnetic ordering in UO_2 corresponds to the irreducible star k_{10} (in the notation of Ref. 7) with rays

$$\mathbf{k}_1 = {}^1/_2(\mathbf{b}_2+\mathbf{b}_3),\ \mathbf{k}_2 = {}^1/_2(\mathbf{b}_1+\mathbf{b}_3),\ \mathbf{k}_3 = {}^1/_2(\mathbf{b}_1+\mathbf{b}_2), \quad (1)$$

where $\mathbf{b}_1$, $\mathbf{b}_2$, and $\mathbf{b}_3$ are the reciprocal-lattice vectors. The rays $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ are parallel to the x, y, and z axes of the Cartesian frame of the figure.

Following Refs. 8 and 9, we introduce the ferro- and antiferromagnetism vectors

$$\begin{aligned} &\mathbf{F}=\mathbf{S}_1+\mathbf{S}_2+\mathbf{S}_3+\mathbf{S}_4 \quad (\mathbf{k}=0),\\ &\mathbf{L}_1=\mathbf{S}_1+\mathbf{S}_2-\mathbf{S}_3-\mathbf{S}_4 \quad (\mathbf{k}=\mathbf{k}_1),\\ &\mathbf{L}_2=\mathbf{S}_1-\mathbf{S}_2+\mathbf{S}_3-\mathbf{S}_4 \quad (\mathbf{k}=\mathbf{k}_2),\\ &\mathbf{L}_3=\mathbf{S}_1-\mathbf{S}_2-\mathbf{S}_3+\mathbf{S}_4 \quad (\mathbf{k}=\mathbf{k}_3), \end{aligned} \quad (2)$$

the wave number $\mathbf{k}$ in the parentheses determines the translation symmetry of the corresponding vector.

The rest of the analysis will be carried out in the quasiclassical approximation and for temperatures $T \ll T_N$. We can use therefore the normalization conditions

$$\mathbf{S}_1^2=\mathbf{S}_2^2=\mathbf{S}_3^2=\mathbf{S}_4^2\equiv S^2$$

or, in equivalent form [with (2) taken into account],

$$\begin{aligned} &\mathbf{L}_1^2+\mathbf{L}_2^2+\mathbf{L}_3^2+\mathbf{F}^2=(4S)^2,\\ &\mathbf{L}_1\mathbf{L}_2+\mathbf{L}_3\mathbf{F}=\mathbf{L}_1\mathbf{L}_3+\mathbf{L}_2\mathbf{F}=\mathbf{L}_2\mathbf{L}_3+\mathbf{L}_1\mathbf{F}=0. \end{aligned} \quad (3)$$

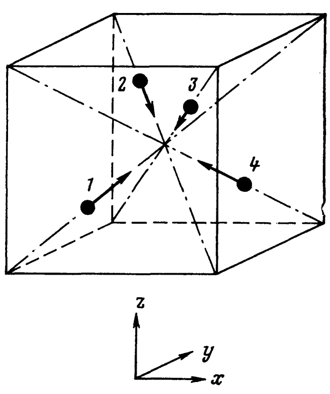

FIG. 1. Magnetic structure of UO_2. The ion magnetic moments are parallel to the body diagonals of the cube.

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The components of the vectors F and L_p ($p = 1, 2, 3$) are distributed over the irreducible representations of the paraphase symmetry group as follows[8]:

$$L_{1x},\ L_{2y},\ L_{3z}, \tag{4}$$

$$L_{1y},\ L_{1z},\ L_{2z},\ L_{2x},\ L_{3x},\ L_{3y}, \tag{5}$$

$$F_x,\ F_y,\ F_z. \tag{6}$$

From the standpoint of exchange symmetry,[1)] the representations (4) and (5) pertain to one permutation representation (exchange multiplet).

The magnetic ordering in UO_2 corresponds to the irreducible representation (4). In this case

$$L_{1x}=L_{2y}=L_{3z}\equiv L_0=4S/3^{1/2}. \tag{7}$$

The magnetic-symmetry group of such a state is $Pn3m'$. The corresponding primitive cell is shown in Fig. 1 and contains four crystal-chemistry cells.

Taking into account the transformation properties of the vectors $\mathbf{F}$ and $\mathbf{L}_p$, as well as the normalization conditions (3), we can obtain the following expressions for the homogeneous part of the exchange energy[8,16]:

$$\mathscr{H}_m^{(e)}={}^1/_2B\mathbf{F}^2+{}^1/_4D(\mathbf{L}_1^4+\mathbf{L}_2^4+\mathbf{L}_3^4). \tag{8}$$

We have left out of (8) the terms biquadratic in spin:

$$\mathbf{F}^4,\ (\mathbf{FL}_1)^2+(\mathbf{FL}_2)^2+(\mathbf{FL}_3)^2,$$

since, as shown in Ref. 16, they are of no significance for the investigation of the static and dynamic properties of UO_2 in the absence of an external field.

In the exchange approximation, the ground state is determined[8] by the parameters B and D of (8). The constant B in UO_2 is large and positive, meaning that in the ground state

$$\mathbf{F}=0,\ \mathbf{L}^2=(4S)^2. \tag{9}$$

The type of ferromagnetic ordering is determined then by the sign of the constant D in (8).

At $D<0$ we have

$$\mathbf{L}_1=\mathbf{L}_2=0;\ L_3=L=4S. \tag{10}$$

At $D>0$,

$$L_1=L_2=L_3\equiv L_0=L/\sqrt{3},\quad \mathbf{L}_1\perp\mathbf{L}_2\perp\mathbf{L}_3\perp\mathbf{L}_1. \tag{11}$$

The orientation of the magnetic structure relative to the crystallographic axes is determined by the relativistic interactions. To within terms quadratic in the spins, we have[8]

$$\mathscr{H}_a=-{}^1/_2a(L_{1x}^2+L_{2y}^2+L_{3z}^2). \tag{12}$$

In the case of UO_2 we must put $a>0$, and have according to (10)–(12)

for phase 1 ($D<0$)

$$\mathbf{L}_1=\mathbf{L}_2=0;\ L_{3z}=L=4S; \tag{13}$$

for phase 2 ($D>0$)

$$L_{1x}=L_{2y}=L_{3z}\equiv L_0=4S/\sqrt{3}. \tag{14}$$

In UO_2 phase 2 is observed (see Fig. 1). We point out once more the distinguishing feature of the magnetic ordering in UO_2. In the exchange approximation, the relative orientation of the sublattice magnetizations is uniquely established only with account taken of the relatively weak biquadratic exchange. This gives rise to singularities in the spectrum of the exchange (optical) magnon mode (Ref. 9) and also to exchange-striction anomalies of the magnetoelastic properties.

MAGNETOELASTIC INTERACTIONS IN UO_2

The magnetostriction quadratic in the spin is determined in the exchange approximation, with allowance for the normalization conditions (3), by the two terms

$$\mathscr{H}_{\rm me}^{(e)}=-\frac{\Lambda}{2}(\mathbf{L}_1^2e_1+\mathbf{L}_2^2e_2+\mathbf{L}_3^2e_3)-\frac{\Lambda_1}{2}\mathbf{F}^2e, \tag{15}$$

where $e=e_1+e_2+e_3$ is the trace of the strain tensor. We use for the components $u_{\alpha\beta}$ of the strain tensor the Voight notation

$$e_1=u_{xx},\ e_2=u_{yy},\ e_3=u_{zz},$$
$$e_4=2u_{yz},\ e_5=2u_{xz},\ e_6=2u_{xy}.$$

The elastic-strain energy density for cubic-syngony crystals is

$$\mathscr{H}_{\rm el}=\frac{c_{11}}{2}(e_1^2+e_2^2+e_3^2)+c_{12}(e_1e_2+e_1e_3+e_2e_3)+\frac{c_{44}}{2}(e_4^2+e_5^2+e_6^2) \tag{16}$$

or, equivalently,

$$\mathscr{H}_{\rm el}=\frac{K}{2}e^2+\frac{c}{2}(\xi^2+\zeta^2)+\frac{c_{44}}{2}(e_4^2+e_5^2+e_6^2), \tag{17}$$

where

$$K=\frac{1}{3}(c_{11}+2c_{12}),\qquad c=c_{11}-c_{12}, \tag{18}$$

$$\xi=\frac{1}{\sqrt{6}}(3e_3-e),\qquad \zeta=\frac{1}{\sqrt{2}}(e_1-e_2). \tag{19}$$

The quantities e, $\{\xi,\zeta\}$, and $\{e_4, e_5, e_6\}$, respectively, constitute the basis of irreducible representations A_{1g}, E_g, and F_{2g} of the point group O_h

From the thermodynamic viewpoint, allowance for $\mathscr{H}_{\rm me}^{(e)}$ [Eq. (15)] is responsible for two significant effects, viz., exchange-striction renormalization of the elastic modulus c of (17) and of the biquadratic exchange constant D. In fact, from the condition that

$$\mathscr{H}^{(e)}=\mathscr{H}_{\rm m}^{(e)}+\mathscr{H}_{\rm me}^{(e)}+\mathscr{H}_{\rm el} \tag{20}$$

be a minimum with respect to the strains e_n at fixed directions of the lattice anharmonicities we get

$$\langle e_n\rangle=\begin{cases}\dfrac{\Lambda}{2c}L_p^2-\dfrac{\Lambda c_{12}}{6cK}L^2+\dfrac{\Lambda_1}{6K}F^2 & (n=p=1,2,3)\\ 0 & (n=4,5,6).\end{cases} \tag{21}$$

Substituting (21) in (20), we obtain the static renormalization of the biquadratic-exchange parameters, and in particular

$$D^*=D-\Lambda^2/2c. \quad (22)$$

We note that if phases 1 and 2 are stable [see Eqs. (10), (11) and (13), (14)] D must also be replaced by D^* [Eq. (22)]. Exchange magnetostriction can thus lead to the onset of the collinear phase 1 even at $D>0$.

On the other hand, from the condition that $\mathscr{H}^{(e)}$ be a minimum with respect to $\mathbf{F}$ and $\mathbf{L}_p$ at fixed strains e_n we obtain in the noncollinear phase

$$F=0, \quad \langle L_p^2\rangle=\frac{1}{3}\left\{(4S)^2+\frac{\Lambda}{D}(3e_n-e)\right\}, \qquad n=p=1,2,3. \quad (23)$$

Substituting (23) in $\mathscr{H}^{(e)}$ of (20), we obtain the static renormalization of the elastic moduli in phase 2, due to the exchange magnetostriction

$$c_{11}^*=c_{11}-\Lambda^2/3D, \quad c_{12}^*=c_{12}+\Lambda^2/6D, \quad c_{44}^*=c_{44} \quad (24a)$$

or

$$c^*=c-\Lambda^2/2D, \quad K^*=K, \quad c_{44}^*=c_{44}. \quad (24b)$$

One should expect the renormalizations (22) and (24) to be quite appreciable, since Λ is the constant of the exchange magnetostriction that is quadratic in the spins, and the biquadratic-exchange parameter D is usually not very large. At the very least, it is customarily assumed that

$$D\ll B. \quad (25)$$

There is no exchange-striction renormalization of the elastic moduli in phase 1 (in our approximation).

We consider now relativistic magnetostriction. When account is taken of the normalization conditions (3), the relativistic magnetostriction quadratic in spin is given by

$$\mathscr{H}_{\mathrm{me}}^{(a)}=-\frac{\lambda_1}{2}(L_{1x}^2e_1+L_{2y}^2e_2+L_{3z}^2e_3)-\frac{\lambda_1'}{2}(L_{1x}^2+L_{2y}^2+L_{3z}^2)e$$
$$-\frac{\lambda_2}{2}(L_{1y}L_{1z}e_4+L_{2x}L_{2z}e_5+L_{3x}L_{3y}e_6)-\frac{\lambda_3}{2}(F_x^2e_1+F_y^2e_2+F_z^2e_3)$$
$$-\frac{\lambda_3'}{2}(F_yF_ze_4+F_xF_ze_5+F_xF_ye_6)-\frac{\lambda}{2}\{L_{1x}(L_{1y}e_6+L_{1z}e_5)$$
$$+L_{2y}(L_{2x}e_6+L_{2z}e_4)+L_{3z}(L_{3x}e_5+L_{3y}e_4)\}. \quad (26)$$

In the analysis of the relativistic effects we can confine ourselves to spin-quadratic terms in the Hamiltonian

$$\mathscr{H}=\mathscr{H}_{\mathrm{m}}+\mathscr{H}_{\mathrm{me}}+\mathscr{H}_{\mathrm{el}}. \quad (27)$$

Here

$$\mathscr{H}_{\mathrm{m}}=\frac{B}{2}\mathbf{F}^2-\frac{a}{2}(L_{1x}^2+L_{2y}^2+L_{3z}^2), \quad (28)$$

$$\mathscr{H}_{\mathrm{me}}=\mathscr{H}_{\mathrm{me}}^{(a)}+\mathscr{H}_{\mathrm{me}}^{(e)}. \quad (29)$$

We consider only the magnetic ordering (14) [phase (2)], which is actually observed in UO_2 (a more detailed analysis, including that of phase 1, is given in Ref. 16).

Simple calculation shows that allowance for the first two terms of (26) reduces to renormalization of the anisotropy constant a:

$$a'=a+\frac{\Lambda(4S)^2}{6K}\left(\lambda_1'+\frac{1}{3}\lambda_1\right). \quad (30)$$

The renormalization of the elastic moduli is determined by the last term of (26), which can be rewritten for phase 2, without allowance for anharmonicity, in the form

$$\mathscr{H}_{\mathrm{me}}^{(a)}={}^1/_2\lambda L_0(e_4F_x+e_5F_y+e_6F_z), \quad (31)$$

where account is taken of the fact that the normalization conditions (3) lead to

$$F_x\approx-(L_{2z}+L_{3y}), \quad F_y\approx-(L_{1z}+L_{3x}), \quad F_z\approx-(L_{2x}+L_{1y}).$$

From the condition that $\mathscr{H}$ [Eq. (27)] be a minimum with respect to $\mathbf{F}$ we have at fixed strains e_n, taking (31) into account,

$$F_x=-\frac{\lambda L_0}{2B}e_4, \quad F_y=-\frac{\lambda L_0}{2B}e_5, \quad F_z=-\frac{\lambda L_0}{2B}e_6. \quad (32)$$

These equations determine the linear piezomagnetic effect in UO_2. Eliminating $\mathbf{F}$ from $\mathscr{H}$ with the aid of (32) we obtain the magnetostriction renormalization of the modulus c_{44}:

$$c_{44}^*=c_{44}-\lambda^2L_0^2/4B. \quad (33)$$

This result is somewhat unusual. The point is that the shear modulus is appreciably renormalized in any collinear magnetic structure, viz.,

$$\Delta\mu\sim\lambda^2/H_a, \quad (34)$$

where H_a is the anisotropy field. In particular, vanishing of the anisotropy (in a spin-flip phase transition) is always accompanied for a collinear magnet by vanishing of a certain shear modulus, hence also of the velocity of transverse sound in a definite direction. Nothing of this kind occurs in UO_2 (i.e., in phase 2), where the renormalization is very small and is insensitive to the anisotropy.

As for the renormalization of the moduli c_{11} and c_{12} [see Eqs. (24)], they are quite appreciable in UO_2 and are of exchange-striction origin; this is likewise possible only in a crystal with an exchange-noncollinear magnetic structure.

According to (32), a magnetic field should produce in UO_2 shear strains that are linear in the field. Namely, at $H\ll BL_0$ we have

$$e_4=-\frac{\lambda L_0}{2c_{44}}F_x, \quad e_5=-\frac{\lambda L_0}{2c_{44}}F_y, \quad e_6=-\frac{\lambda L_0}{2c_{44}}F_z, \quad (35)$$

where $F\approx HB^{-1}$. In the immediate vicinity of T_N, where the para-process is significant, the normalization conditions do not hold. The linear piezomagnetic field is then determined by a contribution biquadratic in spin to H_{me}, of the form

$$L_{1x}L_{2y}L_{3z}(F_xe_4+F_ye_5+F_ze_6). \quad (36)$$

SPECTRUM OF COUPLED MAGNETOELASTIC WAVES IN UO_2

We investigate in the sections that follow the dynamics of the coupled magnetic and elastic subsystems at low values of the wave vectors $\mathbf{q}$. We consider only the noncollinear magnetic ordering (14), which is just the one observed in UO_2.

It is shown in Ref. 9 that the magnon spectrum of UO_2 has one exchange (optical) mode and three acoustic ones. The exchange mode corresponds to oscillations of the quantities

$$\tilde{L}_{1x},\ \tilde{L}_{2y},\ \tilde{L}_{3z}, \tag{37}$$

where the tilde indicates the oscillating part of the corresponding projection of the vector $\mathbf{L}_p$ or $\mathbf{F}$. The three acoustic magnons correspond respectively to oscillations of the type

$$1)\ \tilde{F}_x,\ \tilde{L}_{2z},\ \tilde{L}_{3y};\quad 2)\ \tilde{F}_y,\ \tilde{L}_{1z},\ \tilde{L}_{3x};\quad 3)\ \tilde{F}_z,\ \tilde{L}_{1y},\ \tilde{L}_{2x}. \tag{38}$$

In addition, there are three acoustic phonon (sound) modes, which are coupled in the general case with the magnons.

As to the optical phonons, we shall neglect their contribution to the spin dynamics and assume that the corresponding frequencies exceed greatly the spin-system oscillation frequencies.

COUPLING BETWEEN OPTICAL MAGNONS AND SOUND

In this section we use the exchange approximation throughout. In the continual approach, the system Hamiltonian is

$$\hat{H}=\int dV\hat{\mathscr{H}}=\int dv\{\hat{\mathscr{H}}_{\rm m}^{(e)}+\hat{\mathscr{H}}_{\rm me}^{(e)}+\mathscr{H}_{\rm el}\}, \tag{39}$$

where $\hat{\mathscr{H}}_{\rm m}^{(e)}$ and $\hat{\mathscr{H}}_{\rm me}^{(e)}$ are the exchange and exchange-striction contributions to the energy density, and $\mathscr{H}_{\rm el}$ is the elastic-energy density (16). We have

$$\hat{\mathscr{H}}_{\rm m}^{(e)}=\frac{1}{2}B\hat{\mathbf{F}}^2+\frac{1}{4}D(\hat{\mathbf{L}}_1^4+\hat{\mathbf{L}}_2^4+\hat{\mathbf{L}}_3^4)+\frac{\alpha}{2}\sum_{p\beta}\frac{\partial\hat{\mathbf{L}}_p}{\partial x_\beta}\frac{\partial\hat{\mathbf{L}}_p}{\partial x_\beta}+\frac{\beta}{2}\left\{\left(\frac{\partial\hat{\mathbf{L}}_1}{\partial x}\right)^2+\left(\frac{\partial\hat{\mathbf{L}}_2}{\partial y}\right)^2+\left(\frac{\partial\hat{\mathbf{L}}_3}{\partial z}\right)^2\right\}. \tag{40}$$

The last two terms of (40) describe the inhomogeneous exchange interaction. We have left out of (40) the interaction

$$\left(\frac{\partial\hat{\mathbf{F}}}{\partial x}\right)^2+\left(\frac{\partial\hat{\mathbf{F}}}{\partial y}\right)^2+\left(\frac{\partial\hat{\mathbf{F}}}{\partial z}\right)^2. \tag{41}$$

It makes no contribution to the exchange-mode dynamics in UO_2 if there is no external field.

The carets of the symmetrized operators $\hat{\mathbf{F}}$ and $\hat{\mathbf{L}}_p$ in (40) and (41) mean that

$$\mathbf{F}=\langle\hat{\mathbf{F}}\rangle,\quad \mathbf{L}_p=\langle\hat{\mathbf{L}}_p\rangle,\quad p=1,2,3. \tag{42}$$

To investigate the spin-system dynamics we use a method proposed in Ref. 17 and developed in Ref. 16 for the case $\mathbf{q}\neq0$. A brief description of the method and the equations used are given in the Appendix. Using the Hamiltonian (39) and going through the steps (A8)–(A11), taking into account the commutation relations (A13), we obtain the following equations of motion that describe the optical magnon mode with allowance for the exchange magnetostriction in the long-wave region

$$\dot{\tilde{L}}_{1x}=2L_0^3D(\tilde{L}_{2y}-\tilde{L}_{3z})-\alpha L_0\nabla^2(\tilde{L}_{2y}-\tilde{L}_{3z})-\beta L_0\left(\frac{\partial^2\tilde{L}_{2y}}{\partial y^2}-\frac{\partial^2\tilde{L}_{3z}}{\partial z^2}\right)-\Lambda L_0^2(\tilde{u}_{yy}-\tilde{u}_{zz}), \tag{43}$$

where $\tilde{u}_{\alpha\beta}$ is the oscillating part of the strain tensor. The equations of motion for $\tilde{L}_{2y}$ and $\tilde{L}_{3z}$ are obtained from (43) by cyclic permutation of the indices:

$$x\to y\to z\to x;\quad 1\to2\to3\to1. \tag{44}$$

Rather than writing them out, we proceed directly to the Fourier transforms with respect to the temporal and spatial variables. The result is

$$\begin{aligned}&i\omega\tilde{L}_{1x}+Q_y\tilde{L}_{2y}-Q_z\tilde{L}_{3z}-i\Lambda L_0^2(q_y\tilde{u}_y-q_z\tilde{u}_z)=0,\\&i\omega\tilde{L}_{2y}+Q_z\tilde{L}_{3z}-Q_x\tilde{L}_{1x}-i\Lambda L_0^2(q_z\tilde{u}_z-q_x\tilde{u}_x)=0,\\&i\omega\tilde{L}_{3z}+Q_x\tilde{L}_{1x}-Q_y\tilde{L}_{2y}-i\Lambda L_0^2(q_x\tilde{u}_x-q_y\tilde{u}_y)=0,\end{aligned} \tag{45}$$

where

$$Q_\alpha=L_0\{2L_0^2D+\alpha q^2+\beta q_\alpha^2\};\quad \alpha=x,\ y,\ z. \tag{46}$$

Finally, to obtain a closed system of equations we must derive equations of motion for the displacement-vector components $\tilde{u}_x$, $\tilde{u}_y$, and $\tilde{u}_z$. According to Ref. 1 we have

$$\rho\ddot{u}_\alpha=\frac{\partial}{\partial x_\beta}\frac{\partial\mathscr{H}}{\partial u_{\alpha\beta}}=c_{\alpha\beta\mu\nu}\frac{\partial^2u_\mu}{\partial x_\beta\,\partial x_\nu}+\frac{\partial}{\partial x_\beta}\frac{\partial\mathscr{H}_{\rm me}^{(e)}}{\partial u_{\alpha\beta}}. \tag{47}$$

Taking (15) into account and taking the Fourier transforms, we get

$$\begin{aligned}&\rho^{-1}c_{x\beta\mu\nu}q_\beta q_\nu\tilde{u}_\mu-\omega^2\tilde{u}_x+i\Lambda L_0\rho^{-1}q_x\tilde{L}_{1x}=0,\\&\rho^{-1}c_{y\beta\mu\nu}q_\beta q_\nu\tilde{u}_\mu-\omega^2\tilde{u}_y+i\Lambda L_0\rho^{-1}q_y\tilde{L}_{2y}=0,\\&\rho^{-1}c_{z\beta\mu\nu}q_\beta q_\nu\tilde{u}_\mu-\omega^2\tilde{u}_z+i\Lambda L_0\rho^{-1}q_z\tilde{L}_{3z}=0.\end{aligned} \tag{48}$$

The system of Eqs. (45) and (48) describes fully the dynamics of coupled magnetoelastic waves in the exchange approximation for small wave vectors $\mathbf{q}$. It is found that in this approximation the acoustic magnons are coupled neither with sound nor with exchange magnons, so that their analysis can be deferred to the next section, where relativistic effects will also be taken into account.

In the absence of magnetoelastic coupling (at $\Lambda=0$) the dispersion law for dispersion magnons is given, according to (45), by

$$\omega_S^2(\mathbf{q})=Q_xQ_y+Q_xQ_z+Q_yQ_z=L_0^2(2L_0^2D+\alpha q^2+\beta q_x^2)(2L_0^2D+\alpha q^2+\beta q_y^2)+L_0^2(2L_0^2D+\alpha q^2+\beta q_x^2)(2L_0^2D+\alpha q^2+\beta q_z^2)+L_0^2(2L_0^2D+\alpha q^2+\beta q_y^2)(2L_0^2D+\alpha q^2+\beta q_z^2). \tag{49}$$

A similar expression is given in Ref. 9, where it was derived with allowance for nearest-neighbor exchange interaction.

We shall write out a dispersion equation that describes coupled magnetoelastic waves only for two directions of the wave vector, — $\mathbf{q}\|[100]$ and $\mathbf{q}\|[110]$:

1) $\mathbf{q}\|[100]$ $(q_x=q;\ q_y=q_z=0)$.

The optical magnon interacts here only with longitudinal sound. The corresponding dispersion equation is

$$(\omega_l^2-\omega^2)(\omega_S^2-\omega^2)-2\Lambda^2\rho^{-1}L_0^3Qq_x^2=0, \tag{50}$$

where

$$\omega_l=(c_{11}/\rho)^{1/2}q_x\equiv v_lq_x,\quad Q=L_0(2L_0^2D+\alpha q_x^2). \tag{51}$$

The solution of (50) is

$$\omega_\pm={}^1/_2\{\omega_S^2+\omega_l^2\pm[(\omega_S^2-\omega_l^2)^2+8\Lambda^2\rho^{-1}L_0^3Qq_x^2]^{1/2}\}. \tag{52}$$

As $\mathbf{q}\to 0$ the solution $\omega_-(\mathbf{q})$ corresponds to a longitudinal quasiphonon $\omega_-\to\omega_l$ as $\Lambda\to 0$, while $\omega_+(\mathbf{q})$ corresponds to an optical quasimagnon $\omega_+\to\omega_S$ as $\Lambda\to 0$.

It can be easily deduced from (52) that at small $\mathbf{q}\|x$ the longitudinal quasisound velocity is

$$v_l^*=(c_{11}^*/\rho)^{1/2} \quad (\mathbf{q}\|[100]), \tag{53}$$

where c_{11}^* is the effective elastic modulus of the renormalized exchange magnetostriction [see (24)]. The transverse sound, on the other hand, does not interact with the magnetic subsystem and its frequency is determined in the usual manner

$$\omega_{t1}=\omega_{t2}=(c_{44}\rho^{-1})^{1/2}q \quad (\mathbf{q}\|[100]), \tag{54}$$

2) $\mathbf{q}\|[110]$ $(q_x=q_y=q/\sqrt{2};\quad q_z=0)$.

An optical magnon interacts in this case with longitudinal sound and with one of the transverse acoustic modes (with the mode having $\mathbf{u}\perp z$). The corresponding dispersion equation is

$$(\omega_S^2-\omega^2)(\omega_l^2-\omega^2)(\omega_{t1}^2-\omega^2)+\left(\frac{\Lambda^2L_0^3}{2\rho}q^2\right)^2$$
$$-\frac{\Lambda^2L_0^3}{2\rho}q^2\{\omega_l^2(2Q_z+Q_x)+\omega_{t1}^2Q_x-2\omega^2(Q_x+Q_z)\}=0, \tag{55}$$

where

$$\omega_l=[(c_{11}+c_{12}+2c_{44})/2\rho]^{1/2}q\equiv v_lq,$$
$$\omega_{t1}=[(c_{11}-c_{12})/2\rho]^{1/2}q=(c/2\rho)^{1/2}\equiv v_{t1}q. \tag{56}$$

One other transverse acoustic mode (with $\mathbf{u}\|z$) does not interact with the magnetic subsystem; its dispersion law coincides with (54).

The dispersion equation (55) is bicubic. We write down only the solutions that describe two quasiphonon modes at small $\mathbf{q}$:

$$\omega_l^*=(\omega_l^2-\Lambda^2q^2/12\rho D)^{1/2}=v_l^*q, \tag{57}$$

$$\omega_{t1}^*=(\omega_{t1}^2-\Lambda^2q^2/4\rho D)^{1/2}=v_{t1}^*q, \tag{58}$$

where

$$v_l^*=[(c_{11}^*+c_{12}^*+2c_{44}^*)/2\rho]^{1/2},\quad v_{t1}^*=(c^*/2\rho)^{1/2}; \tag{59}$$

here c_{mn}^* are the effective elastic moduli of Eq. (24), renormalized by the exchange magnetostriction.

According to (55), at large $\mathbf{q}$ one can no longer treat longitudinal and transverse quasisound separately [pure transverse sound in the $\mathbf{q}\|[100]$ direction has a polarization $\mathbf{u}\|z$ and a dispersion law (54)].

Let us summarize. In the exchange approximation, only exchange magnons interact with sound in UO_2. This interaction in quite strong, with a coupling parameter proportional to Λ^2D^{-1}.

According to (22) and (14), in the exchange approximation the UO_2 magnetic structure is stable when the parameter $D^*=D-\Lambda^2/2c$ is positive. D^* vanishes at the stability limit of phase 2. It is of interest that the activation of the exchange magnon mode remains finite in this case (even in the exchange approximation!)

$$\omega_S(0)=3^{1/2}L_0^3\Lambda^2c^{-1}\quad \text{at}\quad D^*=0, \tag{60}$$

but on the other hand the quasisound velocity v_{11}^* [Eq. (59)] vanishes in directions that are crystallographically equivalent to [110]. The reason is that the loss of system stability at $D^*=0$ corresponds to a phase transition that according to the symmetry classification is a proper ferroelastic transition even in the exchange approximation.[6] In contrast to the example analyzed in Ref. 6, such a phase transition realizes the passive two-dimensional irreducible representation of the phase-2 symmetry group (an invariant that is cubic in the order parameter is present) and cannot be a second-order phase transition.

ACOUSTIC MAGNONS; ALLOWANCE FOR RELATIVISTIC INTERACTIONS

When the acoustic modes of the magnon spectrum and their coupling with sound are considered, it suffices to retain in the Hamiltonian the terms quadratic in the spins

$$\hat{H}=\int\hat{\mathscr{H}}dV=\int\{\hat{\mathscr{H}}_{\rm m}+\hat{\mathscr{H}}_{\rm me}+\hat{\mathscr{H}}_{\rm el}\}dV, \tag{61}$$

where $\hat{\mathscr{H}}_{\rm el}$ is defined as before by (16) or (17), and $\hat{\mathscr{H}}_{\rm m}$ is given in the approximation quadratic in the spin by

$$\hat{\mathscr{H}}_{\rm m}=\frac{1}{2}B\hat{\mathbf{F}}^2-\frac{1}{2}a(\hat{L}_{1x}^2+\hat{L}_{2y}^2+\hat{L}_{3z}^2)+\frac{\alpha}{2}\sum\frac{\partial\hat{\mathbf{L}}_p}{\partial x_\beta}\frac{\partial\hat{\mathbf{L}}_p}{\partial x_\beta}$$
$$+\frac{\beta}{2}\left\{\left(\frac{\partial\hat{\mathbf{L}}_1}{\partial x}\right)^2+\left(\frac{\partial\hat{\mathbf{L}}_2}{\partial y}\right)^2+\left(\frac{\partial\hat{\mathbf{L}}_3}{\partial z}\right)^2\right\}. \tag{62}$$

We have left out of (62) the exchange term (41) as well as the gradient terms of relativistic origin. These approximations are justified if the exchange interactions are much stronger than the relativistic ones.

For $\hat{\mathscr{H}}_{\rm me}$ in (61) we have ultimately

$$\hat{\mathscr{H}}_{\rm me}=\hat{\mathscr{H}}_{\rm me}^{(e)}+\hat{\mathscr{H}}_{\rm me}^{(a)},$$
$$\hat{\mathscr{H}}_{\rm me}^{(e)}=-\frac{\Lambda}{2}\left(\hat{\mathbf{L}}_1^2\frac{\partial u_x}{\partial x}+\hat{\mathbf{L}}_2^2\frac{\partial u_y}{\partial y}+\hat{\mathbf{L}}_3^2\frac{\partial u_z}{\partial z}\right), \tag{63}$$
$$\hat{\mathscr{H}}_{\rm me}^{(a)}=\frac{\lambda L_0}{2}\left\{\hat{F}_x\left(\frac{\partial u_y}{\partial z}+\frac{\partial u_z}{\partial y}\right)\right.$$
$$\left.+\hat{F}_y\left(\frac{\partial u_x}{\partial z}+\frac{\partial u_z}{\partial x}\right)+\hat{F}_z\left(\frac{\partial u_x}{\partial y}+\frac{\partial u_y}{\partial x}\right)\right\}.$$

We have left out of (63) the terms whose role reduces to the renormalization (30) of the anisotropy constant a. We regard this constant as already renormalized and begin with relations (63) for $\hat{\mathscr{H}}_{\rm me}$.

The subsequent analysis of the magnon-spectrum acoustic modes, with allowance for the magnetoelastic coupling, follows exactly the same procedure as in the case of exchange (optical) magnons (see the preceding section). The pertinent calculation is described in detail in Ref. 16. We present only the results for the spectrum of coupled magnetoelastic waves in the particular case $\mathbf{q}\|[100]$, i.e., for $q=q_x$. This particular case illustrates quite fully the general situation.

At $\mathbf{q}\|x$ the following modes are independent (at small $q=q_z$):

1) $\tilde{F}_x, \tilde{L}_{2z}, \tilde{L}_{3y}$ (acoustic magnon)

$$\omega(\mathbf{q})=[2BL_0^2(a+\alpha q^2)]^{1/2}, \tag{64}$$

2) $\tilde{F}_y, \tilde{L}_{1z}, \tilde{L}_{3x}, \tilde{u}_z$ (acoustic magnon + transverse sound)

$$\omega_\pm(\mathbf{q})=\frac{1}{2}\left\{\omega_{2S}^2+\omega_t^2\pm\left[(\omega_{2S}^2-\omega_t^2)^2+2\lambda^2L_0^4\rho^{-1}\times\left[a+\left(\alpha+\frac{\beta}{2}\right)q^2\right]q^2\right]^{1/2}\right\} \quad (65)$$

where

$$\omega_t=(c_{44}\rho^{-1})^{1/2}q, \quad \omega_{2S}=\left[2BL_0^2\left[a+\left(\alpha+\frac{\beta}{2}\right)q^2\right]\right]^{1/2}, \quad (66)$$

the solutions ω_+ and ω_- correspond as $\mathbf{q}\to 0$ to the acoustic quasimagnon and to the transverse quasisound. The quasisound velocity is then (as $\mathbf{q}\to 0$)

$$v_t^*=[v_t^2-(\lambda^2L_0^2/4B\rho)]^{1/2}=(c_{44}^*/\rho)^{1/2}, \quad (67)$$

where c_{44}^* is defined in (33).

3) $\tilde{F}_z, \tilde{L}_{1y}, \tilde{L}_{2x}, \tilde{u}_y$ (acoustic magnon + transverse sound).

Relations (65)–(67) hold for this case without change.

4) $\tilde{L}_{1x}, \tilde{L}_{2y}, \tilde{L}_{3z}, \tilde{u}_x$ (exchange magnon + longitudinal sound). These modes are described by relations (50)–(53) of the preceding section.

Thus, at $\mathbf{q}\|x$ we have a strong coupling of the longitudinal sound with the exchange magnon and a considerably weaker coupling of the transverse sound with two acoustic magnon modes. The third acoustic magnon mode is not coupled with the sound at all.

The situation is similar in principles for all other directions of the wave vector $\mathbf{q}$, viz., only exchange magnons interact effectively with the sound. We recall once more that in collinear (or in weakly noncollinear) antiferromagnets the situation is the exact opposite—the acoustic magnons interact effectively with the sound, whereas the exchange magnons are hardly coupled with the sound.

APPENDIX

Calculation of the magnon spectrum by the technique of symmetrized spin operators

Group-theoretical methods have long been productive in the calculation of elementary-excitation spectra and in the determination of normal vibrational modes.[18,13] They involve essentially a transition from the ordinary dynamic variables that describe small deviations of a system from equilibrium to linear combinations of these variables, which transform in accordance with irreducible representations of the ground-state symmetry group. In investigating small oscillations of a spin system it is possible, within the standard spin-wave[2)] approximation, to advance farther and use effectively the crystallomagnetic symmetry of the system even before transforming to the operators of small deviations of the spin system from the equilibrium position. We have in mind here the method we proposed previously[17] for determining the normal modes and the corresponding oscillation frequencies of a spin system. There, however, as in all subsequent papers where this method was used (see, e.g., Refs. 6 and 13), only homogeneous oscillations were considered, i.e., modes with $\mathbf{q}=0$. We develop below a natural generalization of this method to include the case when the magnon wave vector $\mathbf{q}$ is in the vicinity of an arbitrary symmetric point of the Brillouin zone. The advantage of the method described below (as well as of its particular realization)[17] is most pronounced in investigations of complex noncollinear magnetic structures.

The starting point is the quantum-mechanical equation of motion for the spin operator of an isolated ion

$$i\frac{\partial}{\partial t}\hat{S}_\alpha(\mathbf{f},\varkappa)=[\hat{S}_\alpha(\mathbf{f},\varkappa),\hat{H}], \quad (A1)$$

where $\alpha=x,y,z$: $\mathbf{f}$ and $\varkappa$ indicate respectively the magnetic-cell coordinate and the magnetic-ion position in this cell; $\hat{H}$ is the system Hamiltonian expressed in terms of spin operators. The spin operators satisfy the commutation relations

$$[\hat{S}_\alpha(\mathbf{f},\varkappa),\ \hat{S}_\beta(\mathbf{f}',\varkappa')]=i\varepsilon_{\alpha\beta\gamma}\hat{S}_\gamma(\mathbf{f},\varkappa)\delta_{\mathbf{ff}'}\delta_{\varkappa\varkappa'}. \quad (A2)$$

In the continual approach we let

$$\hat{S}_\alpha(\mathbf{f},\varkappa)\to v_m\hat{S}_\alpha(\mathbf{r},\varkappa), \quad (A3)$$

where v_m is the magnetic-cell volume; we then obtain in lieu of (A2)

$$[\hat{S}_\alpha(\mathbf{r},\varkappa),\ \hat{S}_\beta(\mathbf{r}',\varkappa')]=i\varepsilon_{\alpha\beta\gamma}\hat{S}_\gamma(\mathbf{r},\varkappa)\delta(\mathbf{r}-\mathbf{r}')\delta_{\varkappa\varkappa'}. \quad (A4)$$

We note further that the analysis of spin excitations in the vicinity of symmetric Brillouin-zone points can be easily reduced to the case of small $\mathbf{q}$, i.e., to an investigation of the vicinity of the point $\mathbf{q}=0$ of the Brillouin zone. It suffices for this purpose to choose not a primitive magnetic cell, but an expanded unit cell such that the corresponding symmetric point of the Brillouin zone is now the center of a new reduced Brillouin zone. The sublattice index $\varkappa$ will now run through a larger number of values, and the number of magnon modes will increase correspondingly. Their identification, however, will encounter no misunderstandings. A similar procedure is used also to investigate the vicinity of the point $\mathbf{q}=0$ of a magnetic Brillouin zone of a structure in which the magnetic and chemical cells do not coincide. In this case the initial cell of the group G_{pm} must be chosen such that all the magnetic structures under investigation not cause its multiplication.

Taking all this into account, we confine ourselves to an investigation of long-wave spin-system excitations. In the continuum approach, the Hamiltonian $\hat{H}$ of the magnetic subsystem is

$$\hat{H}=\int dV\hat{\mathscr{H}}(\mathbf{r}), \quad (A5)$$

where the energy-density operator $\hat{\mathscr{H}}(\mathbf{r})$ is expressed in terms of the spin-density operators of the magnetic sublattices

$$\hat{\mathbf{S}}(\mathbf{r},\varkappa)=v_m^{-1}\hat{\mathbf{S}}(\mathbf{f},\varkappa) \quad (A6)$$

and of their derivatives. Here v_m is the volume of the expanded unit cell.

We transform next to symmetrized operators $\hat{\Gamma}(\mathbf{r})$ in accordance with the scheme

$$\hat{\Gamma}_{nj}^{\mu}(\mathbf{r})=\sum c_{\alpha nj}^{(\mu)}(\varkappa)\hat{S}_\alpha(\mathbf{r},\varkappa), \quad (A7)$$

where the index $\varkappa$ runs through all the values inside the expanded unit cell. Thus, the operators Γ of (A7) effect the irreducible representations of the paraphase symmetry group for the point $\mathbf{q}=0$ of the reduced Brillouin zone.

This transformation must be carried out both in the expression for the density of the Hamiltonian $\mathscr{H}(\mathbf{r})$ (in which case its structure is radically simplified), and in the equations of motion (A1). We get then in place of (A1)

$$i\dot{\hat{\Gamma}}(\mathbf{r})=[\hat{\Gamma}(\mathbf{r}),\hat{H}]=\int dV'[\hat{\Gamma}(\mathbf{r}),\hat{\mathscr{H}}(\mathbf{r}')]. \qquad (A8)$$

The commutation relations for the operators $\hat{\Gamma}(\mathbf{r})$ are

$$[\hat{\Gamma}_{nj}^{(\mu)}(\mathbf{r}),\hat{\Gamma}_{n'j'}^{(\mu')}(\mathbf{r}')]=i\delta(\mathbf{r}-\mathbf{r}')\sum A_{n''j''}^{(\mu'')}\hat{\Gamma}_{n''j''}^{(\mu'')}(\mathbf{r}), \qquad (A9)$$

the mixing coefficients A are determined from (A7) and (A4).

After calculating the commutators in the right-hand side of (A8), the equations of motion are linearized in the spirit of the random-phase approximation; this is equivalent to the substitution

$$\hat{A}\hat{B}\ldots\hat{C}\Rightarrow AB\ldots C+\tilde{A}B\ldots C+A\tilde{B}\ldots C+\ldots+AB\ldots\tilde{C}. \qquad (A10)$$

Here

$$A=\langle\hat{A}\rangle,\ B=\langle\hat{B}\rangle,\ldots,\ C=\langle\hat{C}\rangle,$$
$$\tilde{A}=\hat{A}-A,\ \tilde{B}=\hat{B}-B,\ldots,\ \tilde{C}=\hat{C}-C. \qquad (A11)$$

The terms of the type $AB\ldots C$ in the right-hand side of the equations of motion (A8) cancel out if the ground state (i.e., the mean values) is correctly chosen. The equations of motion are reduced as a result to a system of linear homogeneous equations. This system breaks up automatically into blocks which correspond to oscillations of different symmetry.

A useful relation for the calculation of the commutators in the right-hand side of (A8) and for the subsequent linearization of the equations of motion in accordance with (A10) is

$$\left[\hat{\Gamma}(\mathbf{r}),\frac{\partial}{\partial x'}\hat{A}(\mathbf{r}')\right]\hat{B}(\mathbf{r}')\Rightarrow-[\hat{\Gamma}(\mathbf{r}),\hat{A}(\mathbf{r}')]\frac{\partial}{\partial x}\hat{B}(\mathbf{r}).$$

It was derived using the fact that integration is carried out with respect to the variable $\mathbf{r}'$ [see (A8)].

The method described above enables us to use not only the symmetry of the ground state (i.e., of the magnetic ordering), but also the higher symmetry of the paraphase. The greatest simplification is obtained when the spin Hamiltonian $\hat{H}$ is expressed in terms of the symmetrized operators Γ, and also when the linearization procedure (A10) is used. For noncollinear magnetic structures it becomes unnecessary to introduce a local coordinate frame for each of the magnetic sublattices.

In conclusion, we present commutation relations of type (A9) for the symmetrized operators

$$\begin{aligned}\hat{\mathbf{F}}(\mathbf{r})&=\hat{\mathbf{S}}_1(\mathbf{r})+\hat{\mathbf{S}}_2(\mathbf{r})+\hat{\mathbf{S}}_3(\mathbf{r})+\hat{\mathbf{S}}_4(\mathbf{r}),\\ \hat{\mathbf{L}}_1(\mathbf{r})&=\hat{\mathbf{S}}_1(\mathbf{r})+\hat{\mathbf{S}}_2(\mathbf{r})-\hat{\mathbf{S}}_3(\mathbf{r})-\hat{\mathbf{S}}_4(\mathbf{r}),\\ \hat{\mathbf{L}}_2(\mathbf{r})&=\hat{\mathbf{S}}_1(\mathbf{r})-\hat{\mathbf{S}}_2(\mathbf{r})+\hat{\mathbf{S}}_3(\mathbf{r})-\hat{\mathbf{S}}_4(\mathbf{r}),\\ \hat{\mathbf{L}}_3(\mathbf{r})&=\hat{\mathbf{S}}_1(\mathbf{r})-\hat{\mathbf{S}}_2(\mathbf{r})-\hat{\mathbf{S}}_3(\mathbf{r})+\hat{\mathbf{S}}_4(\mathbf{r}),\end{aligned} \qquad (A12)$$

namely,

$$[\hat{L}_{1\alpha}(\mathbf{r}),\hat{L}_{2\beta}(\mathbf{r}')]=i\varepsilon_{\alpha\beta\gamma}\delta(\mathbf{r}-\mathbf{r}')\hat{L}_{3\gamma}(\mathbf{r}),$$
$$[\hat{L}_{1\alpha}(\mathbf{r}),\hat{L}_{1\beta}(\mathbf{r}')]=[\hat{F}_{\alpha}(\mathbf{r}),\hat{F}_{\beta}(\mathbf{r}')]=i\varepsilon_{\alpha\beta\gamma}\delta(\mathbf{r}-\mathbf{r}')\hat{F}_{\gamma}(\mathbf{r}), \qquad (A13)$$
$$[\hat{F}_{\alpha}(\mathbf{r}),\hat{L}_{1\beta}(\mathbf{r}')]=i\varepsilon_{\alpha\beta\gamma}\delta(\mathbf{r}-\mathbf{r}')\hat{L}_{1\gamma}(\mathbf{r}).$$

The remaining relations are obtained from (A13) by the cyclic permutation $1\to2\to3\to1$ of the indices.

[1)]The necessary exchange-symmetry-theory data are contained in Refs. 10–15. Where possible, we shall adhere to their terminology.

[2)]We consider here systems for which effects connected with one-ion anisotropy are of no significance.

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