Spin waves in UO_2

I. E. Dzyaloshinskiĭ and B. G. Kukarenko

L.D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted 3 July 1978)
Zh. Eksp. Teor. Fiz. **75**, 2290–2294 (December 1978)

The calculation of the spin-wave spectrum in a 4-sublattice antiferromagnet UO_2 is calculated. In accordance with the general premises of exchange symmetry [see, e.g., Halperin and Saslow, Phys. Rev. **B16**, 2154 (1977)], there are three zero-gap modes. In the paired Heisenberg interaction approximation, however, there appears a fourth zero-gap mode with quadratic dependence on the momentum. The gap in this mode is due only to biquadratic exchange. The corresponding heat capacity and magnetic susceptibility are determined.

PACS numbers: 75.30.Ds, 75.30.Et, 75.10.Jm, 75.50.Ee

The antiferromagnet UO_2 is the first known substance consisting of four noncollinear sublattices. From the point of view of symmetry (see, e.g., Ref. 1), such a system is three zero-gap Goldstone modes with a linear dispersion law—spin waves. We shall show, however, that in the simplest form of exchange interaction—Heisenberg quadratic exchange—there is a fourth zero-gap mode with an energy that depends quadratically on the momentum. The gap appears in this mode only because of biquadratic exchange.

The magnetic structure of UO_2 was recently determined experimentally by Faber, Lander, and Cooper[2] (for its description see also Ref. 3). Within the framework of the Landau theory, the question of the possible types of magnetic structures in UO_2 was considered by Man'ko and one of us.[4] The first-order phase transition in UO_2 was subsequently attributed to the influence of fluctuations in the vicinity of the phase-transition point.[5-7] In these theories[4-7] the magnetic structure of UO_2 was determined by the spins

$$\mathbf{S}^1(000),\quad \mathbf{S}^2(0\tfrac{1}{2}\tfrac{1}{2}),\quad \mathbf{S}^3(\tfrac{1}{2}0\tfrac{1}{2}),\quad \mathbf{S}^4(\tfrac{1}{2}\tfrac{1}{2}0)$$

of the four uranium ions in the cubic cell of the crystal or by their linear combinations

$$\mathbf{M}=\mathbf{S}^1+\mathbf{S}^2+\mathbf{S}^3+\mathbf{S}^4,\quad \mathbf{L}_1=\mathbf{S}^1+\mathbf{S}^2-\mathbf{S}^3-\mathbf{S}^4,$$
$$\mathbf{L}_2=\mathbf{S}^1-\mathbf{S}^2+\mathbf{S}^3-\mathbf{S}^4,\quad \mathbf{L}_3=\mathbf{S}^1-\mathbf{S}^2-\mathbf{S}^3+\mathbf{S}^4.$$

Both in the Landau theory and in the theory where account is taken of the fluctuations, two types of magnetic structure turned out to be possible[4-7]: a collinear two-sublattice structure

$$\mathbf{L}_1\neq 0,\quad \mathbf{L}_2=\mathbf{L}_3=\mathbf{M}=0 \tag{I}$$

and a noncollinear four-sublattice structure

$$L_1=L_2=L_3,\quad \mathbf{L}_1\perp\mathbf{L}_2\perp\mathbf{L}_3,\quad \mathbf{M}=0. \tag{II}$$

For the magnetic structure of type (II) we calculate below the spin-wave spectrum at $T=0$ and obtain expressions for the heat capacity and for the magnetic susceptibility in the limit of large ion spins ($S\gg 1$).

In the classical limit, the exchange decreases exponentially with distance, and we confine ourselves therefore in the Hamiltonian to the following terms:

$$\begin{aligned}
\mathcal{H}=&\frac{1}{2}\sum \mathcal{J}_{ij}(\mathbf{S}_i^1\mathbf{S}_j^1+\mathbf{S}_i^2\mathbf{S}_j^2+\mathbf{S}_i^3\mathbf{S}_j^3+\mathbf{S}_i^4\mathbf{S}_j^4)\\
&+\sum J_{ij}(\mathbf{S}_i^1\mathbf{S}_j^2+\mathbf{S}_i^1\mathbf{S}_j^3+\mathbf{S}_i^1\mathbf{S}_j^4+\mathbf{S}_i^2\mathbf{S}_j^3+\mathbf{S}_i^2\mathbf{S}_j^4+\mathbf{S}_i^3\mathbf{S}_j^4)\\
&-\sum a_{ij}(S_{ix}^1S_{jx}^2+S_{ix}^3S_{jx}^4+S_{iy}^1S_{jy}^3+S_{iy}^2S_{jy}^4+S_{iz}^1S_{jz}^4+S_{iz}^2S_{jz}^3)\\
&+\frac{1}{2}\sum I_{ij}\{(\mathbf{S}_i^1\mathbf{S}_j^2)^2+(\mathbf{S}_i^1\mathbf{S}_j^3)^2+(\mathbf{S}_i^1\mathbf{S}_j^4)^2+(\mathbf{S}_i^2\mathbf{S}_j^3)^2+(\mathbf{S}_i^2\mathbf{S}_j^4)^2+(\mathbf{S}_i^3\mathbf{S}_j^4)^2\}\\
&+\sum K_{ijkl}\{(\mathbf{S}_i^1\mathbf{S}_j^2)(\mathbf{S}_k^3\mathbf{S}_l^4)+(\mathbf{S}_i^1\mathbf{S}_j^3)(\mathbf{S}_k^2\mathbf{S}_l^4)+(\mathbf{S}_i^1\mathbf{S}_j^4)(\mathbf{S}_k^2\mathbf{S}_l^3)\}\\
&+\sum E_{ikl}\{(\mathbf{S}_i^1\mathbf{S}_k^2)(\mathbf{S}_i^1\mathbf{S}_l^3)+(\mathbf{S}_i^1\mathbf{S}_k^2)(\mathbf{S}_i^1\mathbf{S}_l^4)+(\mathbf{S}_i^1\mathbf{S}_k^3)(\mathbf{S}_i^1\mathbf{S}_l^4)\\
&+(\mathbf{S}_i^2\mathbf{S}_k^1)(\mathbf{S}_i^2\mathbf{S}_l^3)+(\mathbf{S}_i^2\mathbf{S}_k^1)(\mathbf{S}_i^2\mathbf{S}_l^4)+(\mathbf{S}_i^2\mathbf{S}_k^3)(\mathbf{S}_i^2\mathbf{S}_l^4)\\
&+(\mathbf{S}_i^3\mathbf{S}_k^1)(\mathbf{S}_i^3\mathbf{S}_l^2)+(\mathbf{S}_i^3\mathbf{S}_k^1)(\mathbf{S}_i^3\mathbf{S}_l^4)+(\mathbf{S}_i^3\mathbf{S}_k^2)(\mathbf{S}_i^3\mathbf{S}_l^4)\\
&+(\mathbf{S}_i^4\mathbf{S}_k^1)(\mathbf{S}_i^4\mathbf{S}_l^2)+(\mathbf{S}_i^4\mathbf{S}_k^1)(\mathbf{S}_i^4\mathbf{S}_l^3)+(\mathbf{S}_i^4\mathbf{S}_k^2)(\mathbf{S}_i^4\mathbf{S}_l^3)\}\\
&-g\mu_B\mathbf{H}\sum(\mathbf{S}_i^1+\mathbf{S}_i^2+\mathbf{S}_i^3+\mathbf{S}_i^4).
\end{aligned} \tag{1}$$

Here $\mathcal{J}_{ij}$ and J_{ij} are the exchange integrals; a_{ij} are the anistropy constants; I_{ij}, K_{ijkl}, and E_{ikl} are the biquadratic exchange integrals; H is the magnetic field; g is the gyromagnetic ratio; μ_B is the Bohr magneton. All the sums in the Hamiltonian are taken over the nearest neighbors.

In the nearest-neighbor approximation, the Fourier components of the exchange integrals are given by

$$J^{12}=2J\left(\cos\frac{k_y+k_z}{2}+\cos\frac{k_y-k_z}{2}\right),\quad J^{13}=2J\left(\cos\frac{k_x+k_z}{2}+\cos\frac{k_x-k_z}{2}\right),$$
$$J^{14}=2J\left(\cos\frac{k_x+k_y}{2}+\cos\frac{k_x-k_y}{2}\right), \tag{2}$$

where the indices ij and J designate the numbers of the sublattices,

$$\mathcal{J}_{\mathbf{k}}=2\mathcal{J}(\cos k_x+\cos k_y+\cos k_z);$$

$\mathbf{k}$ is measured in units of the reciprocal length of the crystallographic cell.

We assume throughout that $a,\ I,\ K,\ E\ll J,\ |\mathcal{J}|$ and neglect therefore the dependence of the Fourier components a_{ij}, I_{ij}, K_{ijkl}, E_{ikl} on $\mathbf{k}$. At $H=0$, the condition that the energy of the magnetic structure (II) be less

 0038-5646/78/1201155-03$02.40

than the energy of the structure (I) takes the form $I+2K-4E>0$. We assume also that $a>0$. Then the equilibrium state takes the form

$$\mathbf{L}_1=L\mathbf{x},\quad \mathbf{L}_2=L\mathbf{y},\quad \mathbf{L}_3=L\mathbf{z},\quad L=4S/3^{1/2}.$$

In the limit $S\gg 1$, the spin-wave spectrum is determined from the linearized classical equations of motion for the spin, which are conveniently written for $\mathbf{l}_i$ and $\mathbf{m}_i$ ($\mathbf{l}$ and $\mathbf{m}$ are the deviations of $\mathbf{L}$ and $\mathbf{M}$ from equilibrium). At $H=0$ the equations of motion break up into four independent groups: the equations for $l_{1y}l_{2x}m_z$, $l_{1z}l_{3x}m_y$, $l_{2z}l_{3y}m_x$ and $l_{1x}l_{2y}l_{3z}$. The spectrum branches $\omega_i=\omega_i(\mathbf{k})$, $i=1,2,3$, corresponding to the vibrations $l_{1y}l_{2x}m_z$, $l_{1z}l_{3x}m_y$ and $l_{2z}l_{3y}m_x$, form a three-dimensional representation of the cubic group. The frequency of the $l_{1y}l_{2x}m_z$ vibration is

$$\omega_1{}^2={}^1/{}_3S^2\{(\bar J_{\mathbf{k}}-\bar J_0+J_0-J^{14}+8a)^2-(J^{12}-J^{13})^2$$
$$+2(\bar J_{\mathbf{k}}-\bar J_0+J_0-J^{14}+8a)(\bar J_{\mathbf{k}}-\bar J_0+J_0+J^{12}+J^{13}+J^{14})\}. \qquad (3)$$

At small $\mathbf{k}$, the expression for the frequency ω_1 takes the form

$$\omega_1{}^2={}^{32}/{}_3S^2J[8a-\bar Jk^2+{}^1/{}_2J(k_x{}^2+k_y{}^2)]. \qquad (4)$$

In the exchange approximation ($a=0$) the modes ω_i of the spectrum turn out to have zero gap and have the linear dependence on k usually possessed by antiferromagnets. The absence of a gap for the modes ω_i at $a=0$ is due to the fact that the oscillations $l_{1y}l_{2x}m_z$, $l_{1z}l_{3x}m_y$ and $l_{2z}l_{3y}m_x$ at $\mathbf{k}=0$ correspond to rotation of the spin system as a whole. In the absence of anisotropy, such a rotation is free.

From (4) follows the condition for the stability of the magnetic structure, namely $\bar J<0$.

The presence of anisotropy does not influence the oscillation of the order parameter $l_{1x}l_{2y}l_{3z}$. The oscillation $l_{1x}l_{2y}l_{3z}$ corresponds to the frequency

$$\omega^2=\Delta^2+{}^2/{}_3\Delta S\{3(\bar J_{\mathbf{k}}-\bar J_0)+3J_0-J^{12}-J^{13}-J^{14}\}$$
$$+{}^1/{}_3S^2\{3(\bar J_{\mathbf{k}}-\bar J_0)^2+2(\bar J_{\mathbf{k}}-\bar J_0)(3J_0-J^{12}-J^{13}-J^{14})+(J^{12}-J_0)^2$$
$$-(J^{13}-J^{14})^2+(J^{13}-J_0)^2-(J^{12}-J^{14})^2+(J^{14}-J_0)^2-(J^{12}-J^{13})^2\}. \qquad (5)$$

We have put here

$$\Delta={}^{32}/{}_3S^3(I+2K-4E),\quad J_0=\bar J_{\mathbf{k}=0},\quad J_0=J^{12,13,14}_{\mathbf{k}=0} \qquad (6)$$

This branch of the spectrum has an exchange gap Δ determined only by the biquadratic exchange. The relation $\omega=\omega(\mathbf{k})$ has cubic symmetry.

At small $\mathbf{k}$, the expression for the frequency takes the form

$$\omega^2=\Delta^2+{}^2/{}_3\Delta S(J+3|\bar J|)k^2$$
$$+{}^1/{}_3S^2\{(3\bar J^2+2|\bar J|J)k^4+J^2(k_x{}^2k_y{}^2+k_x{}^2k_z{}^2+k_y{}^2k_z{}^2)\}. \qquad (7)$$

At $\Delta=0$ we have $\omega\sim k^2$, i.e., the oscillation of the order parameter corresponds to a spectrum mode with a quadratic dispersion law.

In a magnetic field $\mathbf{H}\parallel\mathbf{z}$, the magnetic structure (II) acquires amagnetic moment $\mathbf{M}\parallel\mathbf{z}$, the vector $\mathbf{L}_1$ acquires a y component $L_{1y}=-M/2$, while the vector $\mathbf{L}_2$ acquires an x component $L_{2x}=-M/2$. Recognizing that a, I, K, $E\ll J$, the expression for the magnetic moment takes the form $M_z=g\mu_BH/J_0$. In this state the $l_{1x}l_{2y}l_{3z}$ oscillation is connected with the $l_{1y}l_{2x}m_z$ oscillation, while $l_{1z}l_{3x}m_y$ is connected with $l_{2z}l_{3y}m_x$. The expression for the frequencies $\tilde\omega_{2,3}$ at small $\mathbf{k}$ becomes ($\tilde\omega_i$ is the frequency corresponding to ω_i in a magnetic field)

$$\tilde\omega_{2,3}{}^2={}^1/{}_2[\omega_2{}^2+\omega_3{}^2+h^2]\pm{}^1/{}_2\{[\omega_2{}^2+\omega_3{}^2+h^2]^2-4\omega_2{}^2\omega_3{}^2\}^{1/2}, \qquad (8)$$

where

$$\omega_2{}^2={}^{32}/{}_3S^2J(8a+|\bar J|k^2+{}^1/{}_2J(k_x{}^2+k_z{}^2)),$$
$$\omega_3{}^2={}^{32}/{}_3S^2J(8a+|\bar J|k^2+{}^1/{}_2J(k_y{}^2+k_z{}^2))$$

are the oscillation frequencies $l_{1z}l_{3x}m_y$ and $l_{2z}l_{3y}m_x$, respectively, at $H=0$; $h=g\mu_BH$.

At $\mathbf{k}=0$ formula (8) yields an expression for the two antiferromagnetic-resonance frequencies

$$\tilde\omega_{2,3}=\pm\frac{h}{2}+\left[\frac{h^2}{4}+\frac{2^8}{3}JaS^2\right]^{1/2}. \qquad (9)$$

The expressions for $\tilde\omega^2$ and $\tilde\omega_1^2$ contain terms $\sim H^2$ which have an additional smallness $(a/J)^2$, $(I/J)^2$, and are too cumbersome to be written out here.

We present now an expression for the heat capacity C at the temperatures $(aJ)^{1/2}$, $\Delta\ll T\ll J$, $H=0$. By virtue of the linear character of the dispersion, the contribution to the heat capacity from the branches $\omega_i(\mathbf{k})$ ($i=1,2,3$) is proportional to T^3. The spectrum branch $\omega(\mathbf{k})$ has a quadratic dispersion, so that its contribution to the heat capacity is $\propto T^{3/2}$:

$$C=\frac{3^{1/2}\pi^2T^3}{160(2)^{1/2}S^3(|\bar J|)^{1/2}J^{3/2}(J+2|\bar J|)}+\frac{5(3)^{7/4}\zeta(5/2)T^{3/2}}{2^5(\pi SJ)^{3/2}}I_1\left(\frac{|\bar J|}{J}\right); \qquad (10)$$

$\zeta(x)$ is a Riemann function.

$$I_1(\alpha)=\int\frac{do}{4\pi}[\alpha(3\alpha+2)+(n_x{}^2n_y{}^2+n_x{}^2n_z{}^2+n_y{}^2n_z{}^2)]^{-3/4};$$

$I_1(\alpha)\approx 1/3^{3/4}\alpha^{3/2}$ at $\alpha\geqslant 5$. The values of $I_1(\alpha)$ at $\alpha<5$ are listed in the table. Just as the heat capacity C, the susceptibility $\chi(H)$ contains at $(aJ)^{1/2}$ and $\Delta\ll T\ll J$ two terms that have different dependences on the temperature. Since $\tilde\omega_{2,3}\sim k$, at $T\gg(aJ)^{1/2}$ the contribution to the susceptibility from the branches $\tilde\omega_{2,3}$ is $\propto T^2$. The integrals corresponding to the contribution made to the susceptibility by the branches $\tilde\omega(\mathbf{k})$ and $\tilde\omega_1(\mathbf{k})$ at $T\gg\Delta$ and $(aJ)^{1/2}$ are determined by the values $k^2\sim\Delta/J$ and a/J, and turn out to be $\propto T$. At arbitrary ratio of the constants Δ and a, we obtain very cumbersome expressions, and we present the answer for only two limiting cases, $a\ll\Delta$ and $\Delta\ll a$:

1) $a\ll\Delta$:

$$\Delta\chi=\frac{3^{1/2}(g\mu_B)^2T^2}{2^8(SJ)^3}I_2\left(\frac{|\bar J|}{J}\right)-T(g\mu_B)^2\frac{2^{1/2}S\Delta^{1/2}(K-E)}{3^{1/2}\pi J^{1/2}}I_3\left(\frac{|\bar J|}{J}\right), \qquad (11)$$

where

TABLE I.

	α									
	0	0,01	0,05	0,1	0,2	0,5	0,7	1	2	5
$I_1(\alpha)$	4.71	3.64	2.54	1.90	1.28	0.60	0.43	0.29	0.07	0.04
$I_2(\alpha)$	1.24	1.09	0.91	0.78	0.58	0.30	0.22	0.15	0.06	0.02
$I_3(\alpha)$	1.57	1.27	0.92	0.70	0.48	0.23	0.16	0.11	0.05	0.01

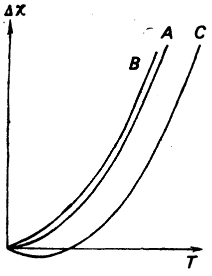

FIG. 1.

$$I_2(\alpha)=\int\frac{do}{4\pi}\{(2\alpha+1-n_x^2)^{1/2}(2\alpha+1-n_y^2)^{1/2}[(2\alpha+1-n_x^2)^{1/2}+(2\alpha+1-n_y^2)^{1/2}]\}^{-1},$$

$$I_3(\alpha)=\int\frac{do}{4\pi}[\alpha(2+3\alpha)+(n_x^2n_y^2+n_x^2n_z^2+n_y^2n_z^2)]^{-1/2}$$
$$\times\{\{1+3\alpha+[1-3(n_x^2n_y^2+n_x^2n_z^2+n_y^2n_z^2)]^{1/2}\}^{1/2}+\{1+3\alpha-[1-3(n_x^2n_y^2+n_x^2n_z^2+n_y^2n_z^2)]^{1/2}\}^{1/2}\}^{-1}.$$

Asymptotically at $\alpha\geqslant 5$ we have

$$I_2(\alpha)\approx 1/4(2)^{1/2}\alpha^{3/2},\quad I_3(\alpha)\approx 1/6\alpha^{3/2}.$$

The values of $I_2(\alpha)$ and $I_3(\alpha)$ at $\alpha<5$ are given in the table. The contribution from the branches $\tilde\omega(\mathbf{k})$ and $\tilde\omega_1(\mathbf{k})$, besides the term given above, contain also a term $\propto T\Delta^{3/2}/J^{7/2}$, which is small compared with the term $\propto T^2/J^3$ in the susceptibility for the temperatures $\Delta\ll T\ll J$.

2) $a\gg\Delta$. In this case the branches $\tilde\omega(\mathbf{k})$ and $\tilde\omega_1(\mathbf{k})$ make a magnetic-susceptibility contribution $\propto Ta^{3/2}/J^{7/2}$. This contribution is small compared with the contribution from the branches $\tilde\omega_2(\mathbf{k})$ and $\tilde\omega_3(\mathbf{k})$ at temperatures $(aJ)^{1/2}\ll T\ll J$ and

$$\Delta\chi=\frac{3^{1/2}(g\mu_B)^2T^2}{2^8(SJ)^3}I_2\left(\frac{|\tilde J|}{J}\right). \tag{12}$$

By virtue of the cubic symmetry of the magnetic structure (II), the expressions obtained for the susceptibility do not depend on the direction of the magnetic field.

A qualitative plot of the function $\Delta\chi=\chi(T)-\chi(0)$ is shown in the figure. The case $a\gg\Delta$ corresponds to the curve A. In the case $a\ll\Delta$, the form of the curve depends on the ratio between the quantities $|K-E|$ and Δ. In the case $|K-E|\ll(J\Delta)^{1/2}$ the qualitative plot of $\Delta\chi=\Delta\chi(T)$ is curve A. The case $|K-E|\gg(J\Delta)^{1/2}$ corresponds to curve B if $K-E<0$ and to curve C if $K-E>0$.

[1]R. I. Halperin and W. M. Saslow, Phys. Rev. B **16**, 2154 (1977).
[2]J. Faber and G. H. Lander, Phys. Rev. B **14**, 1151 (1976); J. Faber, G. H. Lander, and B. R. Cooper, Phys. Rev. Lett. **35**, 1770 (1975).
[3]I. E. Dzyaloshinskiĭ, Commun. Phys. **2**, 69 (1977).
[4]I. E. Dzyaloshinskiĭ and V. Yu. Man'ko, Zh. Eksp. Teor. Fiz. **46**, 1352 (1964) [Sov. Phys. JETP **19**, 915 (1964)].
[5]S. A. Brazovskiĭ, I. E. Dzyaloshinskiĭ, and B. G. Kukharenko, Zh. Eksp. Teor. Fiz. **70**, 2257 (1976) [Sov. Phys. JETP **43**, 1178 (1976)].
[6]V. A. Alessandrini, A. P. Cracknell, and J. A. Przystawa, Commun. Phys. **1**, 51 (1976).
[7]D. Mukamel and S. Krinsky, Phys. Rev. B **13**, 5065, 5078 (1976).

Translated by J. G. Adashko