Spin waves in triple-$\vec{q}$ structures. Application to USb

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The spin-wave spectrum in a system with triple-$\vec{q}$ magnetic structure is calculated. The spin waves differ distinctly from those in the corresponding single-$\vec{q}$ structure, but agree with the excitations observed by Lander and Stirling in uranium antimonide (USb). Their experiments thus directly verify that the spins in USb are ordered in the triple-$\vec{q}$ structure.

A large number of magnetic systems exhibit periodic magnetic structures, which can usually be described by a single wave vector (single-$\vec{q}$ structure). Quite recently, however, it has been suggested that the magnetization in some compounds is formed as superpositions of structures with several symmetric wave vectors (multi-$\vec{q}$ structures).[1-3] Experimentally, it is very difficult to distinguish between a multi-$\vec{q}$ magnetic structure and a system made up of equivalent single-$\vec{q}$ domains. Neutron-diffraction experiments give *identical* patterns in the two cases. An attempt has been made to remove this ambiguity by considering the magnetoelastic effects.[1-3] If the lattice is observed to be distorted in a way which is incompatible with a multi-$\vec{q}$ structure (or with a single-$\vec{q}$ structure), the situation is clear. If the lattice distortion is unobservable, as in UN, UAs, and USb,[4] the ambiguity remains.

In this paper we present calculations of the spin-wave spectrum in the "triple-$\vec{q}$" structure which has been suggested for USb on the basis of a uniaxial-pressure experiment.[3] We find that the spectrum differs distinctly from that of the corresponding single-$\vec{q}$ structure, but agrees with *inelastic* neutron scattering measurements by Lander and Stirling.[5] To be specific, the spectrum includes a *longitudinal* branch not found in single-$\vec{q}$ structures, and a higher-lying transverse branch. These branches become nonaccidentally degenerate at certain reciprocal-lattice points. The spectrum and the stability of the triple-$\vec{q}$ structure can be quantitatively explained by means of a model including anisotropic bilinear couplings and crystal fields only.

Figure 1 shows the triple-$\vec{q}$ and the corresponding single-$\vec{q}$ (type-I) structure in an fcc magnet such as USb. The wave vectors involved are $\vec{q}_z = (2\pi/a)(0,0,1)$, and the two symmetric ones along the x and y directions. Figure 2 is an attempt to show the normal spin-wave modes in the two cases. The modes at the zone boundary X [$\vec{q} = (2\pi/a)(0,0,1)$] are shown. In the single-$\vec{q}$ case the spins in each (001) layer are precessing in phase, but successive layers are 180° out of phase. For other wave vectors along the ΓX direction [$\vec{q} = (2\pi/a)(0,0,q)$] the phase difference differs from 180°. This mode is *transverse* since the polarization of the spin deviations from the equilibrium positions are *perpendicular* to the wave vector (and to the spins).

In the triple-$\vec{q}$ case the situation is more complicated. The spins precess around their equilibrium in a similar way; so the local polarization is perpendicular to the spins. The figure shows two different modes, L and T, given by full vectors and dashed vectors

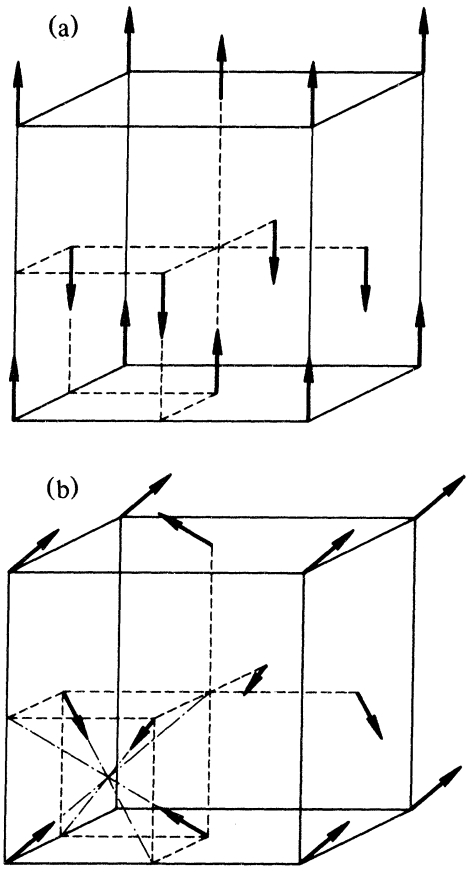

FIG. 1. (a) Single-$\vec{q}$ and (b) triple-$\vec{q}$ magnetic structure in an fcc magnet such as USb.

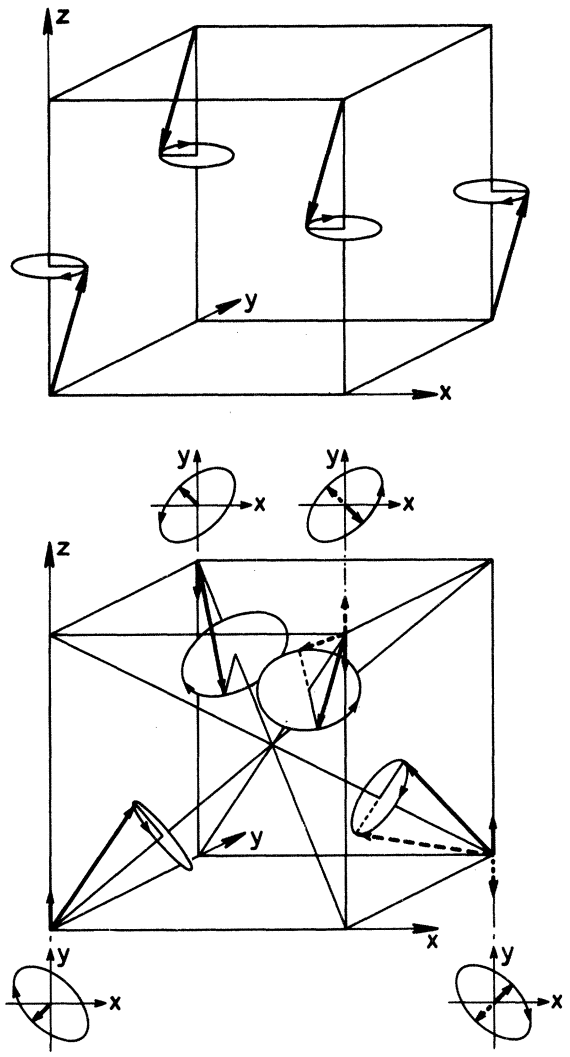

FIG. 2. Normal spin waves in USb. $\vec{q} \| z$ axis. Full lines and vectors: longitudinal mode at X. Dashed lines: transverse mode at X. For comparison, the transverse spin-wave mode in the single-$\vec{q}$ structure is also shown.

(where they differ), respectively. The arrows along the z axis show in a similar notation the projections on the z axis of the precession of the single spins, and the ellipses show the projections on the xy plane. Again the phases between neighbor planes are 180° out of phase. In the L mode the transverse components *within* each layer pairwise cancel each other and cannot be observed in a neutron-diffraction experiment. The longitudinal (z) components, however, are all in phase within each layer. The mode is thus longitudinal and will appear as such in a neutron experiment. This is (as we shall see) the low-lying mode observed by Lander and Stirling (Fig. 3). The second mode, given by the broken vectors, is quite different. The transverse components within each

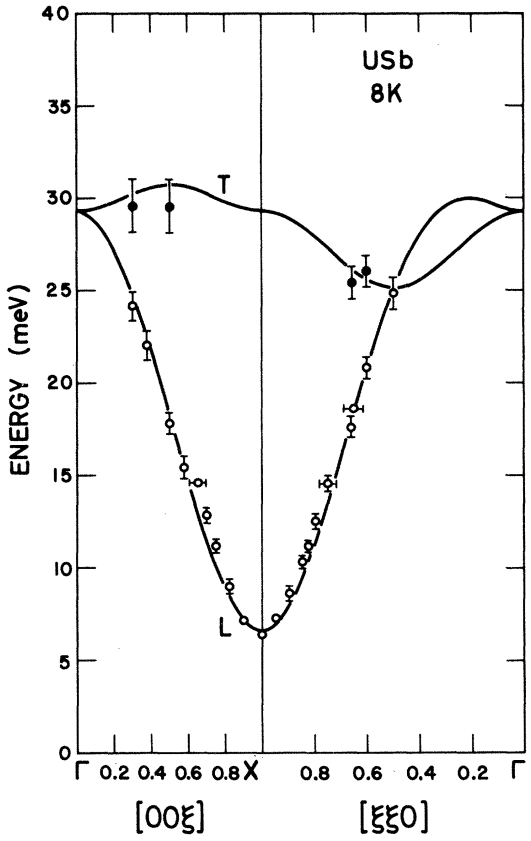

FIG. 3. Spin-wave spectrum in USb. The full lines are calculated as shown in text. The experimental points are taken from Lander and Stirling (Ref. 5). The black points are our own interpretation of their data on the transverse mode.

layer are in phase and the longitudinal components are out of phase, and this mode thus appears as a transverse mode. This is the higher-lying flat mode observed by Lander and Stirling.

To support these general considerations, which are in fact based solely on the symmetry of the structure, we have performed a spin-wave calculation based on a simple model with physically reasonable parameters. This calculation explains quantitatively the experimental spectrum, and the stability of the triple-$\vec{q}$ structure. The Hamiltonian is

$$\begin{aligned} H &= H_{\text{is}} + H_{\text{an}} + H_{\text{cr}} \ , \\ H_{\text{is}} &= -\sum_{nn} J_1 \vec{S}_i \cdot \vec{S}_j \ , \\ H_{\text{an}} &= -\sum_{nn} J_D (\vec{S}_i \cdot \hat{r}_{ij})(\vec{S}_j \cdot \hat{r}_{ij}) \ , \\ H_{\text{cr}} &= B_4^0 (O_4^0 + 5O_4^4) \ , \end{aligned} \tag{1}$$

where the spins are $S = \frac{9}{2}$ (or $S = 4$) corresponding to a $5f^3$ (or $5f^2$) configuration. H_{an} is an anisotropic

pseudodipolar interaction and H_{cr} is a crystal-field term with cubic symmetry. The O_m^n's are fourth-order Stevens operators. Classically, $H_{cr} \sim S_x^4 + S_y^4 + S_z^4$.

The bilinear terms give the same ground-state energy for the single-$\vec{q}$ as for the triple-$\vec{q}$ structure, but the crystal-field term favors the triple-$\vec{q}$ structure for positive B_4^0:

$$E(\text{triple } \vec{q}) - E(\text{single } \vec{q}) = -\tfrac{40}{3} S S_3 B_4^0 \ , \qquad (2)$$

where $S_3 = (S-\frac{1}{2})(S-1)(S-\frac{3}{2})$. The triple-$\vec{q}$ structure is thus stabilized simply by the crystal-field term. This is intuitively clear since in the single-$\vec{q}$ structure all the spins point in the unfavorable [100] direction, whereas the spins in the triple-$\vec{q}$ structure point along [111]. The sign (and size, as we shall see) of B_4^0 agree with estimates of Lander *et al.*[6] based on neutron measurements of the magnetization.

The spin-wave calculation is a standard one based on the Holstein-Primakoff transformation of (1) ($S_{z'} = S - a^+ a$, where z' is the local spin direction). Generally, the spin-wave energies can be written

$$E_{\vec{q}}^2 = A_{\vec{q}}^2 - |B_{\vec{q}}|^2 \ . \qquad (3)$$

Introducing the parameters

$$\begin{aligned} A_0 &= \tfrac{160}{3} S_3 B_4^0 \ , \\ A_1 &= -\tfrac{16}{3} S J_1 - \tfrac{16}{3} S J_D \ , \\ B_1 &= \tfrac{16}{3} S J_1 + \tfrac{4}{3} S J_D \ , \end{aligned} \qquad (4)$$

$A_{\vec{q}}$ and $B_{\vec{q}}$ take the following form for $\vec{q}$ along symmetry directions:

$\vec{q} = (0,0,q)$:

$$L \text{ mode} \begin{cases} A_q = A_0 + A_1[1 - \cos(qa/2)] \\ B_q = B_1[1 - \cos(qa/2)] \ , \end{cases}$$

$$T \text{ mode} \begin{cases} A_q = A_0 + 2A_1 \\ B_q = -B_1[1 + i\sqrt{3}\cos(qa/2)] \ ; \end{cases} \qquad (5a)$$

$\vec{q} = \frac{1}{\sqrt{2}}(q,q,0)$:

$$L \text{ mode} \begin{cases} A_q = A_0 + A_1[\frac{5}{4} - \frac{1}{4}\cos(qa/\sqrt{2}) - \cos(qa/2\sqrt{2})] \\ B_q = B_1[\frac{1}{2} + \frac{1}{2}\cos(qa/\sqrt{2}) - \cos(qa/2\sqrt{2})] \ , \end{cases}$$

$$T \text{ mode} \begin{cases} A_q = A_0 + 2A_1[\frac{7}{8} + \frac{1}{8}\cos(qa/\sqrt{2})] \\ B_q = -B_1[\frac{1}{2} + \frac{1}{2}\cos(qa/\sqrt{2}) + i\sqrt{3}\cos(qa/2\sqrt{2})] \ . \end{cases} \qquad (5b)$$

Figure 3 shows the spin-wave energies calculated for $A_0 = 6.6$ meV, $A_1 = 12.3$ meV, and $|B_1| = 5.4$ meV. The experimental points are taken from Ref. 5. Note the following features:

(i) The dispersion of the longitudinal mode is isotropic near X. This is a consequence of the "cubic" symmetry of the triple-$\vec{q}$ structure. For single-$\vec{q}$ structures the dispersion is not isotropic and, of course, the mode is transverse.

(ii) The two modes are degenerate at the Γ point and at $\vec{q} = (2\pi/a)(\frac{1}{2},\frac{1}{2},0)$. This is again a consequence of the symmetry (nonaccidental).

The intensities of the neutron peaks can also be calculated within the spin-wave theory. For example, the intensities of the two modes are roughly the same in a transverse scan ($\vec{\kappa} \perp \vec{q}$), whereas in a longitudinal scan the intensity of the longitudinal mode vanishes, in agreement with Ref. 5, Fig. 7.

These conclusions do not depend crucially upon the specific model chosen for USb. The spin-wave approximation is adequate since the magnetization is more than 85% of the saturation value and we can neglect single-spin longitudinal response. The parameters deduced are effective ones.

However, the mean-field transition temperature calculated on the basis of A_1 agrees with the actual T_N within a factor 2. The gap at X is a measure of the stability of the triple-$\vec{q}$ structure. In our model, both the gap and the stability [Eq. (2)] are given by the crystal-field term. The crystal-field parameters V_4 and V_6 deduced by Lander *et al.*[6] from form-factor measurements yield $A_0 \sim 10$ meV. $|B_1|$ is not well determined by the dispersion relations and the sign is unknown. The anisotropic interaction $-J_D$ is at least a factor of 2 larger than $|J_1|$. If we choose $B_1 < 0$, J_D and J_1 are both negative and the structure

is stable. Otherwise, next-nearest-neighbor interactions are necessary to stabilize the structure with respect to a single-$\vec{q}$ type-II antiferromagnet with $\vec{S} \| \vec{q} = (\pi/a)(1,1,1)$. The importance of J_D is also indicated both by the absence of transverse scattering,[7] and by the very anisotropic longitudinal spin correlations just above T_N.

We have also considered the effect of the Coqblin-Schrieffer (CS) interaction deduced by Siemann and Cooper.[8] We find that their spin potential is equivalent with the coupling

$$J_{CS}[(S_i^+)^3(S_j^-)^3 + (S_i^-)^3(S_j^+)^3]_{\vec{r}_i \| \vec{r}_{ij}}$$

plus a minor quadrupole-quadrupole interaction. This does not lead to any qualitative changes of the spin-wave model, except that J_{CS} would also be involved in the stability criterion.

In conclusion, we have explained the excitations measured by Lander and Stirling using a simple phenomenological model. The spins in USb order in the triple-$\vec{q}$ structure, and the excitations are spin waves in this structure.

ACKNOWLEDGMENT

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