The group G_q equals D_2 (containing all the translations). The band representations of D_2 are given by formula (11), where $D^{(r,l)}$ are now the representations of the point group D_2. According to Ref. 5, there are two inequivalent points (points f and e on page 179 of Ref. 5) with the symmetry D_2. The corresponding band representations of D_4^1 can be constructed according to the rule given in (7). It can be checked that by starting with the D_4- and D_2-symmetry points we obtain all the irreducible band representations of D_4^1.

In summary, this Letter gives for the first time a symmetry specification of bands in solids based entirely on band representations of space groups. These are new representations of groups which correspond to a band of energies rather than to a single energy as in the case of usual representations. While the symmetry of an energy level in an atom is specified with respect to a single center, the symmetry of a band in a solid is specified with respect to an infinite lattice of centers. This is in full correspondence with the concept of a band in a solid as derived from atomic levels of atoms placed on a lattice. The lattice of the symmetry centers is an invariant property of the band and if the full information about a band becomes accessible it should be possible to determine experimentally the position and the type of the lattice for each band in a solid.

[1]F. Bloch, Z. Phys. 52, 555 (1928).
[2]E. E. Koch, C. Kunz, and B. Sonntag, Phys. Rep. 29, 153 (1977).
[3]G. F. Koster, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1957), Vol. 5.
[4]J. Des Cloizeaux, Phys. Rev. 129, 554 (1963), and 135, A685, A698 (1964).
[5]*International Tables of X-Ray Crystallography*, edited by N. F. M. Henry and K. Lonsdale (Kynoch, Birmingham, 1952), Vol. 1.
[6]G. F. Koster, Phys. Rev. 89, 67 (1953); G. Parzen, Phys. Rev. 89, 237 (1953).
[7]P. W. Anderson, Phys. Rev. Lett. 21, 13 (1968).
[8]J. Zak, Phys. Rev. B 18, 749 (1978).

Magnetic Phase Transition near a Lifshitz Point: A Neutron Study of UAs

S. K. Sinha and G. H. Lander
Argonne National Laboratory, Argonne, Illinois 60439

and

S. M. Shapiro
Brookhaven National Laboratory, Upton, New York 11973

and

O. Vogt
Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule, CH-8093 Zürich, Switzerland
(Received 2 July 1980)

Peculiar rodlike diffuse critical scattering above the type-I antiferromagnetic (AF/I) phase transition has been observed in the actinide compound UAs and demonstrates that the transition is in the vicinity of a Lifshitz point. A mean-field theory is proposed based on an anisotropic Hamiltonian and provides a reasonable description of the results.

PACS numbers: 75.40.Fa, 75.25.+z, 75.50.Ee, 64.60.Kw

Recently there has been considerable interest in phase transitions associated with a so-called Lifshitz point, i.e., a multicritical point separating a uniformly ordered phase, a modulated phase, and a disordered phase.[1] In this Letter we present evidence to show that this criterion is almost satisfied for UAs near its antiferromagnetic ordering temperature. The critical scattering in UAs is anisotropic, exhibits a maximum at an incommensurate point in reciprocal space, and disappears when a superlattice peak appears at the commensurate wave vector. We analyze the critical behavior by developing a mean-field treatment of a Hamiltonian that includes strong cubic anisotropy.

The uranium monopnictides (all with the NaCl crystal structure) provide examples of a family of compounds with strong cubic anisotropy in the

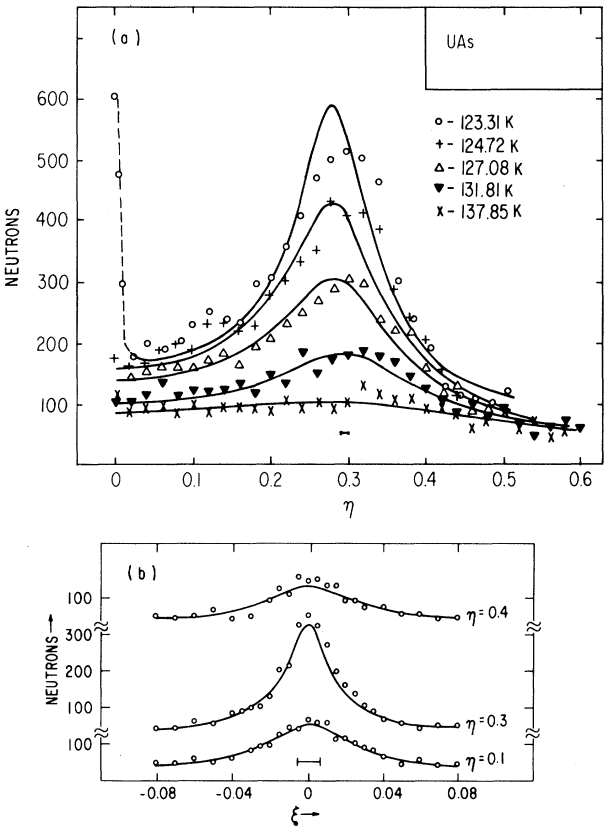

FIG. 1. (a) Intensity along $[00\eta]$ from $\vec{Q} = (2\pi/a)(1,1,0)$ as a function of temperature and (b) along $[\xi\xi 0]$ as a function of η for $T = 124.75$ K. In both figures the points represent experimental observations and the solid lines are theoretical fits. The width of the resolution function is marked by a horizontal bar. Different counting times have been used for (a) and (b).

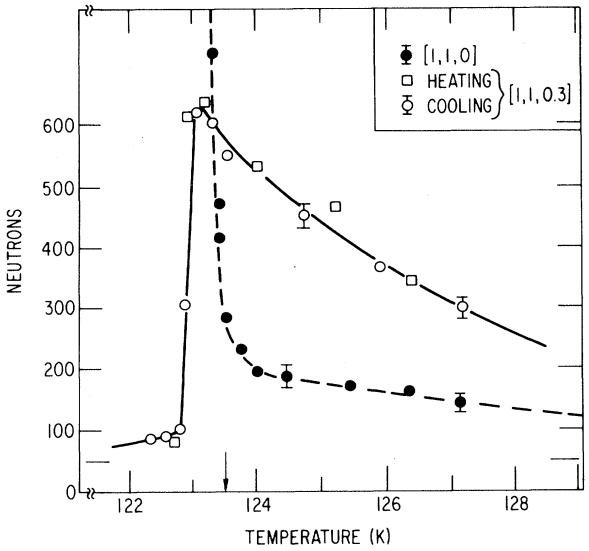

FIG. 2. Intensity as a function of temperature for $\vec{Q} = (2\pi/a)(1,1,0)$ [$\vec{q} = (2\pi/a)(0,0,1)$] and $\vec{Q} = (2\pi/a)(1,1,0.3)$ [$\vec{q} = (2\pi/a)(0,0,0.7)$] positions in UAs. The background level is ~ 50 counts.

magnetic interactions between the U atoms. The systems UN and USb undergo a second-order phase transition to a type-I antiferromagnetic (AF/I) structure.[2,3] In the AF/I phase, the U spins in an [001]-type domain align in (001) sheets with alternating sheets having spins parallel and antiparallel to the [001] axis. Uranium arsenide ($a = 5.768$ Å) also undergoes a AF/I transition at $T_N = 123.5$ K but the transition is weakly first order. At a lower temperature (63.5 K) in UAs another first-order phase transition occurs to the type-IA structure, in which the stacking of (001) sheets is ++−− rather than +−+−.

The sample used was a single crystal of UAs of volume 0.034 cm^3, oriented with the scattering in the $(1\bar{1}0)$ plane. The experiment was carried out on a triple-axis neutron spectrometer at the high-flux-beam reactor at Brookhaven National Laboratory with use of a beam of 13.5-meV neutrons and a 13-cm-thick pyrolytic-graphite filter to remove higher-order contamination of the beam. The study of the diffuse critical scattering was carried out both as a function of wave-vector and energy transfer, but in the latter case no inelasticity was detected within the instrumental energy resolution (0.4 meV), so that we may treat the diffuse scattering as if it were integrated over energy transfer.

Figure 1(a) shows the temperature evolution of the diffuse scattering as measured along the $[00\eta]$ direction. The scattering is much broader than the resolution, and as $T_N = 123.5$ K is approached the intensity has a maximum near $\eta = 0.3$. Figure 1(b) shows some of the scans in the $[\xi\xi 0]$ direction as a function of η for $T = 124.75$ K. The intensity distribution is much narrower in the $[\xi\xi 0]$ than in the $[00\eta]$ direction and thus resembles a cigar elongated along [001]. Another interesting feature of the data, which is reproduced by the fits discussed below, is that the half-width is narrowest when the intensity is greatest. As in USb and UN, no critical scattering was observed around (001) so we conclude that for $\vec{q}$ along [001] only $\chi^{zz}(\vec{q})$ shows critical behavior. Figure 2 shows another unique feature of this critical scattering. As $T \to T_N$, the intensity at $\vec{Q} = (2\pi/a)(1,1,0.3)$ begins to diverge as if the material would like to order at $\vec{q} = (2\pi/a)(0,0,0.7)$ as measured from the (111) zone center, but, before the long-range ordering at this wave vector can

occur, the transition to an AF/I structure occurs with the superlattice peaks appearing at $\vec{q} = (2\pi/a)(0,0,1)$. If we denote the AF superlattice point as $\vec{q}_0 = (2\pi/a)(0,0,1)$ and deviations parallel and perpendicular to $\vec{q}_0$ as $q_\parallel$ and $q_\perp$, respectively, then Fig. 1 shows that $\chi^{zz}(\vec{q})$ is much narrower as a function of $q_\perp$ than along $q_\parallel$, implying longer-range correlations within the (001) sheets than between the sheets.

All of the above behavior may be explained qualitatively in terms of a mean-field treatment of the following effective spin Hamiltonian:

$$H = -\sum_{i,j,\alpha}{}' J^{\alpha\alpha}(\vec{R}_{ij}) S_i{}^{\alpha} S_j{}^{\alpha} - v \sum_{i,\alpha} (S_i{}^{\alpha})^4, \qquad (1)$$

where the anisotropic exchange interaction $J^{\alpha\alpha}(\vec{R}_{ij})$ transforms with $\vec{R}_{ij}$ according to full cubic symmetry, $S_i{}^{\alpha}$ is the α component of the spin operator for the ith ion, and v is a constant associated with the single-ion anisotropy. As we shall see, the present system is consistent with $J^{zz}(\vec{R}) \gg J^{xx}(\vec{R})$ and $J^{yy}(\vec{R})$ for $\vec{R}$ lying in the x-y plane, etc. We assume diagonal interactions between first- and second-neighbor uranium moments on the NaCl lattice, given by

$$J^{zz}(\vec{R}_1) = J_0, \quad J^{xx}(\vec{R}_1) = J^{yy}(\vec{R}_1) = J_1 \; [\vec{R}_1 = (\tfrac{1}{2}a, \tfrac{1}{2}a, 0)];$$
$$J^{zz}(\vec{R}_2) = J_2, \quad J^{xx}(\vec{R}_2) = J^{yy}(\vec{R}_2) = J_3 \; [\vec{R}_2 = (0,0,a)].$$

To preserve symmetry, the tensor components of the interactions must be permuted in an appropriate fashion as $\vec{R}_1$ and $\vec{R}_2$ are taken over the first and second sets of neighbors, respectively. Then a mean-field treatment for the paramagnetic phase yields

$$\chi^{zz}(\vec{q}) = C[T - J^{zz}(\vec{q})CV/g^2\mu_B{}^2]^{-1}, \qquad (2)$$

where g is the Landé factor for the U ions, V the crystal volume, C is the Curie constant, and

$$\begin{aligned} J^{zz}(\vec{q}) = {} & \tfrac{1}{3}vS(S+1) + 2J_0\{\cos[\tfrac{1}{2}(q_x+q_y)a] + \cos[\tfrac{1}{2}(q_x-q_y)a]\} \\ & + 2J_1\{\cos[\tfrac{1}{2}(q_x+q_z)a] + \cos[\tfrac{1}{2}(q_x-q_z)a] + \cos[\tfrac{1}{2}(q_y+q_z)a] + \cos[\tfrac{1}{2}(q_y-q_z)a]\} \\ & + 2J_2\cos(q_x a) + 2J_3[\cos(q_x a) + \cos(q_y a)]. \end{aligned} \qquad (3)$$

We assume that $J_0 > 0$, and $J_1, J_2 < 0$ yielding a maximum for $J^{zz}(\vec{q})$ in the vicinity of $\vec{q}_0 = (2\pi/a)(0,0,1)$. Note that $J^{xx}(\vec{q})$ would be given by Eq. (3) with an appropriate permutation of q_x, q_y, q_z, etc. Evaluation of this expression shows that at $\vec{q}_0$, $J^{xx}(\vec{q})$ and $J^{yy}(\vec{q})$ are very much smaller than $J^{zz}(\vec{q})$, provided that J_0 is the dominant interaction. This explains why critical scattering is seen only from $\chi^{zz}(\vec{q})$ for $\vec{q}$ along [001], although cubic symmetry would, of course, require $\chi^{xx}(\vec{q})$ and $\chi^{yy}(\vec{q})$ to diverge along [100] and [010], respectively. Since the critical scattering is well localized around $\vec{q}_0$ in the $q_\perp$ direction, we may expand to $O(q_\perp{}^2)$ and obtain

$$\chi^{zz}(\vec{q}) = a_1\{1 + [a_2 - a_3\cos(\tfrac{1}{2}aq_\parallel)]a^2q_\perp{}^2/4 + a_4[1 - \cos(aq_\parallel)] - 4a_3[1 - \cos(\tfrac{1}{2}aq_\parallel)]\}^{-1}, \qquad (4)$$

where the mean-field predictions for the temperature dependence of the coefficients a_i are

$$\begin{aligned} a_1 &= C/(T - T_0), \\ a_2 &= (2CV/g^2\mu_B{}^2)(J_0 + 2J_3)/(T - T_0), \\ a_3 &= (2CV/g^2\mu_B{}^2)J_1/(T - T_0), \\ a_4 &= (2CV/g^2\mu_B{}^2)J_2/(T - T_0), \end{aligned} \qquad (5)$$

and T_0 is the mean-field ordering temperature. Note that if $a_4 \simeq a_3$ (implying $J_1 \simeq J_2$), the denominator of $\chi^{zz}(\vec{q})$ would have a vanishing coefficient of $q_\parallel{}^2$ around $\vec{q}_0$. This would correspond to a generalized Lifshitz point where there is a balance between competing antiferromagnetic interactions between first- and second-neighbor (001) sheets of spins and thus a balance between tendencies to sinusoidal and commensurate AF/I ordering.

For the present case, and in the high-temperature limit, the energy integrated neutron diffuse scattering at $\vec{Q} = (2\pi/a)(1,1,0)$ is related to the susceptibility by[4]

$$d\sigma/d\Omega = Af^2(\vec{Q})\chi^{zz}(q), \qquad (6)$$

where A is a constant and $f(\vec{Q})$ is the magnetic form factor of the U ion. With use of the full mean-field expression for $\chi^{zz}(\vec{q})$ in Eq. (4), the expression (6) was folded with the experimental instrumental resolution in momentum space and fitted to the data with use of the parameters a_1, a_2, a_3, and a_4 in Eq. (4) and a constant background. Figure 1 shows the quality of the fits obtained using the mean-field theory at various temperatures and the observed behavior is reproduced quite well. The poorer fit along the η axis indicates a need to include interactions between further neighbor (001) sheets. From the fit, we obtain a $T = 116.2 \pm 0.2$ K, which is below the actual T_N(123.5 K) of the first-order phase transi-

tion. The ratio J_1/J_2 varies between 0.62 and 0.69 as compared to the value 1.0 as the condition for a Lifshitz point. If we assume $J_0/J_1 = J_2/J_3$, then the average value of this ratio is 37.4. This shows that an extremely large cubic anisotropy exists, so that the diffuse scattering would be elongated along $q_\parallel$ even if the competition between J_1 and J_2 did not occur. This extremely large anisotropy, which was also observed in both UN (Ref. 2) and USb (Ref. 3), may be related microscopically to the covalent bonding of the f orbitals on the U atoms with anion p orbitals in (001) sheets. A modified Coqblin-Schrieffer interaction[5] has also been proposed to account for the large anisotropy.

The Hamiltonian invoked for this system has many close analogies with the Ising models studied recently.[6] These results indicate that away from the Lifshitz point the transition from paramagnetic to sinusoidal or from paramagnetic to commensurate AF/I is a second-order transition, but the transition between the sinusoidal and commensurate phases is first order. Interestingly, a AF/IA structure is predicted to be stable at the lowest temperature, as is observed in UAs.

The present system, which shows sinusoidal fluctuations and then enters the commensurate AF/I phase from the paramagnetic phase in a first-order manner, can be accounted for by the use of a Landau expansion of the free energy when the expansion parameters are $q_\parallel, q_\perp$, and a single component of the magnetization. Terms up to sixth order in S_i^α are required to explain the observed behavior. However, a more satisfactory approach must be sought within the applications of renormalization-group theory. The Hamiltonian with cubic anisotropy has been discussed by Aharony[7] and Bruce.[8] For $n = 3$, the transition can be second order with Heisenberg-like critical behavior, although the "irrelevant" cubic anisotropy terms are believed to represent very slowly decaying corrections to the leading scaling behavior. Fluctuations can drive the transition first order in the case of strong cubic symmetry or in the vicinity of a Lifshitz point, even though mean-field theory would predict a second-order transition. USb and UN exhibit second-order paramagnetic-AF/I transitions and display anisotropy comparable to the present case. A corresponding mean-field treatment for USb would yield the value 49.0 for the ratio J_0/J_1 if only first-neighbor interactions are included. Thus the first-order nature of the transition in UAs is due to the additional complication of being in the vicinity of a Lifshitz point.[1]

The magnetic ordering in UAs is analogous to the first-order structural phase transitions in some perovskite-structure compounds.[9,10] Aharony and Bruce[11] have shown that a uniaxial stress can cause such a transition to become second order and thus pass through a tricritical Lifshitz point, for which they have calculated critical exponents and these seem to be confirmed by electron-paramagnetic-resonance measurements on stressed $RbCaF_3$.[10] Further investigations of the detailed critical behavior of UAs are in progress.

We would like to acknowledge useful discussions with A. Aharony, P. Bak, P. A. Lindgard, and G. Shirane. This work was supported by the U. S. Department of Energy under Contracts No. DE-AC02-76CH00016 and No. W-31-109-ENG-38.

[1]R. M. Hornreich, M. Luban, and S. Shtrikman, Phys. Rev. Lett. 35, 1678 (1975); S. Redner and H. E. Stanley, Phys. Rev. B 16, 4901 (1977).

[2]W. J. L. Buyers, T. M. Holden, E. C. Svensson, and G. H. Lander, in *Proceedings of the International Symposium on Neutron Inelastic Scattering, Vienna, 1977* (IAEA, Vienna, 1978), p. 239.

[3]G. H. Lander, S. K. Sinha, D. M. Sparlin, and O. Vogt, Phys. Rev. Lett. 40, 523 (1978).

[4]See, for example, R. M. White, *Quantum Theory of Magnetism* (McGraw-Hill, New York, 1970), Chap. 8.

[5]R. Seimann and B. R. Cooper, Phys. Rev. Lett. 44, 1015 (1980).

[6]W. Selke and M. E. Fisher, Phys. Rev. B 20, 257 (1979); J. van Boehm and P. Bak, Phys. Rev. Lett. 42, 122 (1979).

[7]A. Aharony, Phys. Rev. Lett. 31, 1494 (1973).

[8]A. D. Bruce, J. Phys. C 7, 208 (1974).

[9]G. Shirane, V. J. Minkiewicz, and A. Linz, Solid State Commun. 26, 583 (1978).

[10]J. Y. Buzare, J. C. Fayet, W. Berlinger, and K. A. Muller, Phys. Rev. Lett. 42, 465 (1979).

[11]A. Aharony and A. D. Bruce, Phys. Rev. Lett. 42, 462 (1979).