Incommensurate magnetic order in UPtGe

R. A. Robinson and A. C. Lawson
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

J. W. Lynn*
Reactor Radiation Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899

K. H. J. Buschow
Philips Research Laboratories, 5600 JA Eindhoven, The Netherlands
(Received 3 February 1992; revised manuscript received 24 July 1992)

The orthorhombic uranium (1:1:1) ternary compound UPtGe has been studied by means of neutron powder diffraction, using both the time-of-flight method at a spallation source and the constant-wavelength reactor method. The material exhibits incommensurate magnetic order with $\mathbf{q}=(0,0.5543,0)$, with a uranium moment at the lowest temperature of $1.11\mu_B$ and a Néel temperature of 51 K. The moments are confined to the *bc* plane, which includes the propagation vector for the incommensurate modulation, and they rotate within that plane as one moves parallel to the *b* axis. The magnitude of the ordered uranium moment is the only order parameter in the problem.

I. INTRODUCTION

The ternary (1:1:1) uranium-based compounds U*T*Ge, where *T* is a transition-metal element, exist in the $CeCu_2$ structure type, or its ordered form, the NiSiTi structure type. They have been studied extensively by means of magnetic susceptibility, electrical resistivity, and specific heat.[1,2] Furthermore, magnetization measurements have been made in quasicontinuous fields up to 35 T and pulsed fields up to 50 T.[1,3] In going from T=Co,Ru, which are nonmagnetic, through T=Rh, which is a weak ferromagnet, to T=Pd,Ni,Pt, which appear to exhibit complicated types of 5*f*-moment ordering, one proceeds from itinerant to localized behavior, presumably as a function of the gradual reduction in 5*f*-*d* hybridization.

In this paper we report results of a neutron powder diffraction study of UPtGe. It was reported[1,2] to order antiferromagnetically at 51 K and to have field-induced transitions at 23 and 42 T. We therefore made initial zero-field measurements on the Los Alamos spallation source at 75 and 15 K. We had hoped to determine whether UPtGe crystallized in the NiSiTi structure type, with space group *Pnma*, as reported for UlrGe and UIrSi by Chevalier *et al.*[4] or in the disordered $CeCu_2$ structure type with space group *Imma*. However, on the basis of our data alone we could not distinguish between these two possibilities. Nevertheless, we determined that it exhibited incommensurate magnetic order. These initial experiments were continued at the National Institute of Standards and Technology research reactor with a detailed temperature-dependent study of two of the magnetic reflections. We found that the wavelength of the modulation was temperature independent, leaving only the magnitude of the uranium moment as the order parameter.

II. EXPERIMENTAL METHOD

The sample was prepared by arc melting the constituent elements of at least 99.9% purity under argon gas. After arc melting, the sample was wrapped in Ta foil and vacuum annealed at 800 °C for 16 days. 8.0 g of sample were ground and enclosed in a sealed vanadium tube. This was mounted in a computer-controlled closed-cycle refrigerator. At Los Alamos we used the HIPD diffractometer.[5,6] This instrument has detectors at $\pm153°$, $\pm90°$, and $\pm40°$. All six banks were used to refine the crystallographic structure, while the 40° banks are the most useful for the study of magnetism. At NIST we used the BT-9 triple-axis spectrometer in two-axis mode with a vertically focusing pyrolytic-graphite monochromator and graphite filter. The wavelength was 2.3505 Å. The analyzer had been removed and the collimation was 40′/30′/40′. The relatively coarse resolution was chosen to maximize the integrated intensity.

III. RESULTS

The 75 K data refine fairly well to both the $CeCu_2$ and the NiSiTi structure types with the parameters listed in Table I. Because the numerical values of the scattering lengths of Ge and Pt are rather close, the diffraction patterns give the appearance of a disordered compound, independent of the actual state of atomic order. We were therefore unable to distinguish between the background and the extra reflections present in *Pnma* by eye. However, these reflections have been observed in a recent single-crystal study,[7] and we therefore prefer the NiSiTi-type structure with space group *Pnma*. In what follows we will assume this space group and its coordinate system. The quality of the fit to our power data is not as good as we would like, and we also have some differences between data taken in different banks. We attribute these differences to imperfect particle statistics, because there are not enough grains to achieve a complete powder average. We do not believe that these small variations significantly affect the values of the refined parameters in Table I.

In Fig. 1, we show portions of the 40° data taken on

TABLE I. Refined structural parameters for UPtGe at 75 K.

Space group		Pnma		
U	4c	$x\ \frac{1}{4}\ z$	$x=0.0032\pm0.0003$	$z=0.2158\pm0.0001$
Pt	4c	$x\ \frac{1}{4}\ z$	$x=0.2104\pm0.0002$	$z=0.5907\pm0.0001$
Ge	4c	$x\ \frac{1}{4}\ z$	$x=0.7839\pm0.0002$	$z=0.5813\pm0.0001$

$a=7.1712\pm0.0005$ Å
$b=4.3198\pm0.0003$ Å
$c=7.4993\pm0.0006$ Å

R factors
$R_{wp}=6.14\%$
$R_p=4.51\%$
Reduced $\chi^2=7.886$

HIPD at 15 and 75 K respectively, as well as the difference. At 15 K, a number of extra peaks appear, most notably at d spacings of approximately 5.9 and 7.8 Å. We assume these to be magnetic. They do not occur at d spacings which are simply related to the crystallographic cell, but can be indexed assuming a wave vector of $\mathbf{q}=(0,q,0)$, in other words, with a modulation of some kind along the orthorhombic b-axis. By fitting to the observed d-spacings, listed in Table II, we obtain the value of $q=0.5543\pm0.0001$ in reduced units. Note that we choose to define q in the second Brillouin zone, for the good reason that the systematic absences are then exactly the same as for the crystallographic space group. It would be more conventional to define the wave vector in the first Brillouin zone, but the systematic absences of the magnetic reflection set would then not be easily related to those of the crystallographic space group. For instance, the two strongest peaks are the $000^{\pm}$ and 011^{-} reflections at 7.8 and 5.9 Å, respectively, while the $001^{\pm}$ and $001^{\pm}$ reflections which would occur at 5.3 and 5.4 Å are missing. The simplest type of magnetic ordering to give these absences is one in which the uranium atoms are aligned ferromagnetically within the xz planes but rotated by an angle ϕ between planes, where $\phi=\pi q=99.8°$. One possibility is a helical structure, but the moments could also rotate within a plane other than that perpendicular to $\mathbf{q}$. A moment-density wave with this wave vector would also give these absences. In fact, the most general case to give these systematic absences would be a structure in which the Cartesian components of the moments on the ith uranium atom are

$$\mu_{x,i}=\mu_x\cos(\mathbf{q}\cdot\mathbf{r}_i+\beta)\ ,\mu_{y,i}=\mu_y\cos(\mathbf{q}\cdot\mathbf{r}_i)\ ,$$
$$\mu_{z,i}=\mu_z\cos(\mathbf{q}\cdot\mathbf{r}_i+\alpha)\ , \tag{1}$$

where α and β are phase angles. This model includes, among other things, moment-density waves, a helical structure with moments confined to the ac plane, and other structures in which the moments rotate as one moves along the b direction. Generalizing the standard structure factor for helical magnetic structures,[8] the interference part of the cross section is proportional to

$$I=\left[\frac{(1-\hat{Q}_x^2)}{4}\mu_x^2+\frac{(1-\hat{Q}_y 2)}{4}\mu_y^2+\frac{(1-\hat{Q}_z 2)}{4}\mu_z^2\right.$$
$$-\hat{Q}_x\hat{Q}_y\mu_x\mu_y\cos\beta$$
$$\left.-\hat{Q}_y\hat{Q}_z\mu_{\mu_z}\cos\alpha-\hat{Q}_z\hat{Q}_x\mu_z\mu_x\cos(\alpha-\beta)\right]$$
$$\times\sum_{rr'}\exp(i\tau\cdot(\mathbf{r}-\mathbf{r}')) \tag{2}$$

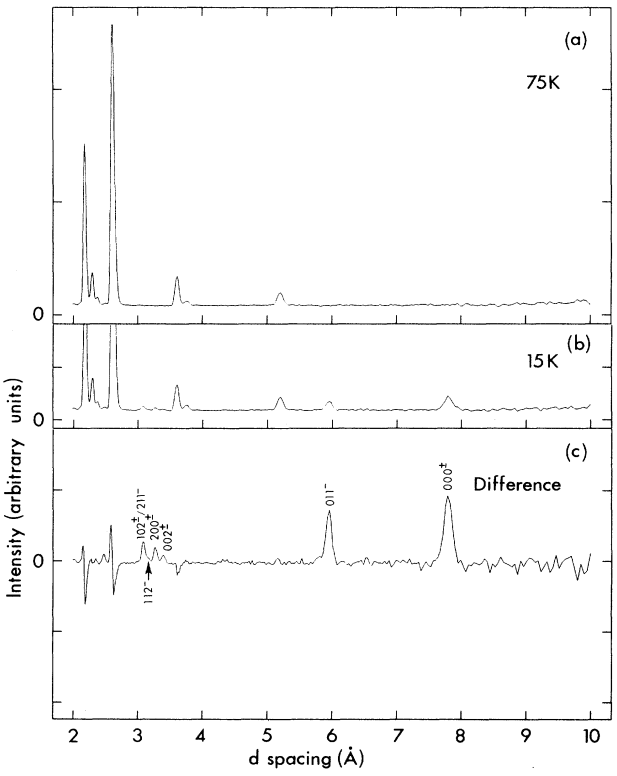

FIG. 1. Plots of a portion of the raw data taken at 40° on HIPD at LANSCE: (a) at 75 K, (b) at 15 K, and (c) the difference between the 15 K data and the 75 K data. The indices in the lower panel are for the low-order magnetic reflections only. The intensities have been divided by the incident spectrum.

TABLE II. Observed magnetic reflections for UPtGe at 15 K.

Indices	d spacing (Å)	Observed cross section (barns/unit cell)	Calculated cross section for model shown in Fig. 2 (barns/unit cell)
$000^{\pm}$	7.781±0.003	0.588±0.019	0.665
000^{-}	5.940±0.002	1.063±0.054	1.249
$002^{\pm}$	3.381±0.001	3.653±0.035	3.713
$200^{\pm}$	3.260±0.001	0.772±0.051	0.473
112^{-}	3.148±0.002	2.270±0.041	2.100
$102^{\pm}/211^{-}$	3.073±0.004	1.067±0.023	0.977
$013^{-}/202^{\pm}$	2.464±0.001	2.748±0.165	2.892

TABLE III. Fitted parameters for magnetic structure of UPtGe at 15 K.

General model	Cycloidal model shown in Fig. 2	General moment-density wave model
$q=0.5543\pm0.0001$	$q=0.5543\pm0.001$	$q=0.5543\pm0.0001$
$\mu_x=0.177\pm0.007\mu_B$ $\mu_y=1.415\pm0.006\mu_B$ $\mu_z=1.024\pm0.008\mu_B$	$\mu=1.109\pm0.004\mu_B$	$\mu_x=0.508\pm0.013\mu_B$ $\mu_y=2.101\pm0.014\mu_B$ $\mu_z=0.721\pm0.012\mu_B$
$\alpha=77.1\pm0.4°$ $\beta=38.2\pm7.3°$		$\alpha=0$ $\beta=0$
Reduced $\chi^2=3.55$	Reduced $\chi^2=3.78$	Reduced $\chi^2=9.2$

where $\hat{Q}$ is a unit vector parallel to $\mathbf{Q}=\tau\pm\mathbf{q},\tau$ being a reciprocal-lattice vector for the crystallographic lattice. If we least-squares fit our observed intensities to this model we obtain the parameters given in Table III. Note that μ_x is much smaller than μ_y and μ_z. This means that the moments are probably confined to the yz plane and that the angle β is irrelevant. Furthermore, we find that the angle α is close to 90°, and we note that if $\alpha=90°$ and $\mu_y=\mu_z$, then the magnetic moments on all the uranium sites would have the same magnitude. This structure is shown in Fig. 2; the moments lie within the bc plane and rotate as y increases from plane to plane by the angular ϕ. The parameters obtained by fitting to this structure, the "cycloidal" structure obtained by Kawamata *et al.*[7] in their single-crystal study, are also listed in Table III. The fit is almost as good and we find this model more appealing on physical grounds, because all uranium atoms carry the same moment and the magnetic structure is far more symmetric. The general model is not that dissimilar, but requires a net amplitude modulation of the uranium moment. In the cycloidal structure, we obtain a moment of $1.109\pm0.004\mu_B$. For purposes of comparison, we also include in Table III the general moment-density wave model, in which all uranium moments are parallel: it is not supported by the data.

In all fits, we have corrected for the Lorentz factor and have assumed the uranium form factor of Freeman *et al.*[9] The magnetic intensities were normalized to both the 101 and 200 nuclear peaks, which lie in the same d-spacing range as the magnetic reflections and fit quite well in the Rietveld refinement of the crystallographic structure. These intensities are listed in absolute units in Table II, along with corresponding cross sections for the cycloidal structure. As we have some overlapping reflections, neither are corrected for powder multiplicities.

At NIST, we studied the temperature dependence of the $000^{\pm}$ and 011^{-} magnetic reflections in more detail. First, neither peak moves in position or broadens significantly with temperature. Second, the ratio of their integrated intensities does not change with temperature.

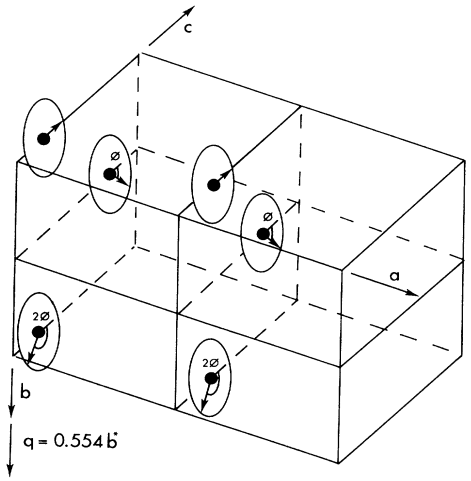

FIG. 2. The magnetic structure of UPtGe. The moments are confined to the bc plane and rotate by an angle ϕ between each layer of uranium moments. Only one crystallographic cell is shown, with four uranium atoms in that cell. The other lines are merely added to help the reader's perspective. For the sake of clarity, only the uranium atoms are shown.

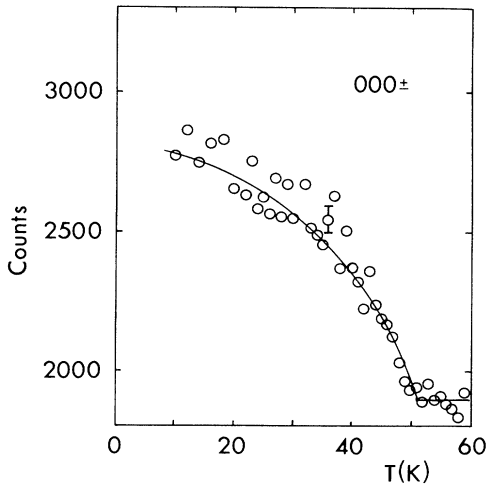

FIG. 3. The variation in intensity of the 000± magnetic reflection, as recorded on BT-9 at NIST. The line is merely a guide to the eye.

It is therefore reasonable to measure the peak intensity of one peak, as a function of temperature, by setting the spectrometer up on the top of the peak and merely changing temperature. In Fig. 3 we show the data taken on the $000^{\pm}$ reflection in this manner. Clearly, the Néel temperature is very close to 51 K, in good agreement with the bulk measurements.[1,2] The intensity is proportional to the square of the uranium moment, which is the only order parameter.

IV. DISCUSSION

Our results and those of Kawamata *et al.*[7], clearly indicate that the *bc* plane is the easy magnetization plane in this compound. Indeed this is exactly what has been seen in a high-field magnetization study[10] on a single crystal of UPtGe: the susceptibility in the *a* direction is lower than in the other two directions. Furthermore, there are metamagnetic transitions at approximately 25 T if the field is in the *bc* plane, while no transition occurs at fields below 30 T if the field is parallel to the *a* axis.

A further interesting point is that the temperature variation of the order parameter shown in Fig. 3 would seem to indicate that the transition is second order. However, it is well known[11] that, in orthorhombic structures, a second order transition should result in a one-dimensional order parameter, namely a moment-density wave like that described in Table III. Such models are clearly inconsistent with our data. It would therefore be worthwhile to investigate the order of the transition more carefully by means of single-crystal diffraction.

We note that similar incommensurate structures have been observed in isostructural rare-earth based compounds TbRhSi and ErRhSi[12] and TbRhGe[13], while HoRhSi[14] is commensurate but noncollinear and HoNiSi[14] is both commensurate and collinear. Likewise, UNiGe and UPdGe are both commensurate and collinear at low temperature, but UPdGe exhibits a moment-density wave between 28 and 50 K.[7]

V. SUMMARY

We have shown that UPtGe exhibits incommensurate magnetic order with $\mathbf{q}=(0,0.5543,0)$ and a uranium moment at the lowest temperature of $1.109\mu_B$. The moments are confined to the *bc* plane but rotate by 99.8° between adjacent planes of uranium atoms. There is only one order parameter, the uranium moment, and the Néel temperature is 51 K. Neither $\mathbf{q}$ nor the widths of the reflections change with temperature. In the course of this study, we became aware of the results of a single-crystal neutron-diffraction study of the same compound by Kawamata *et al.*[7] These authors obtain a uranium moment of $1.0\mu_B$, with the moments lying within the *bc* plane and with $\mathbf{q}=(0,0.57,0)$, which is in good agreement with our results.

ACKNOWLEDGMENTS

We are grateful to V. Sechovsky, L. Havela, and F. R. de Boer for useful discussions and in particular, for drawing our attention to Ref. 7. We are also indebted to J. A. Goldstone for the use of her program FIT_PEAKS, to R. B. Von Dreele for assistance with the Rietveld refinement, and to S. Skanthakumar for help with the experiment at NIST. We are grateful to C. Gonzales and F. Valdez for their assistance with the manuscript. This work was supported in part by the Division of Basic Energy Sciences of the U.S. Department of Energy.

*Also at the University of Maryland, College Park, MD 20742.

[1] R. Troć and V. H. Tran, J. Magn. Magn. Mater. **73**, 389 (1988).
[2] K. H. J. Buschow, E. Brück, R. G. van Wierst, F. R. de Boer, L. Havela, V. Sechovsky, P. Nozar, E. Sugiura, M. Ono, M. Date, and A. Yamagishi, J. Appl. Phys. **67**, 5125 (1990).
[3] F. R. de Boer, E. Brück, V. Sechovsky, L. Havela, and K. H. J. Buschow, Physica B **163**, 175 (1990).
[4] B. Chevalier, B. Lloret, P. Gravereau, B. Buffat and J. Étorneau, J. Magn. Magn. Mater **75**, 13 (1988).
[5] D. K. Hyer, editor, Los Alamos Report No. LALP 90-7, 1990 (unpublished).
[6] A. C. Lawson, J. A. Goldstone, J. G. Huber, A. L. Giorgi, J. W. Conant, A. Severing, B. Cort, and R. A. Robinson, J. Appl. Phys. **69**, 5112 (1991).
[7] S. Kawamata, K. Ishimoto, Y. Yamaguchi, and T. Komatsubara, J. Magn. Magn. Mater, **104-107**, 51 (1992).
[8] G. L. Squires, *Introduction to the Theory of Thermal Neutron Scattering* (Cambridge University, Cambridge, England, 1978), p. 155.
[9] A. J. Freeman, J. P. Desclaux, G. H. Lander, and J. Faber, Phys. Rev. B **13**, 1168 (1976).
[10] S. Kawamata, G. Kido, K. Ishimoto, Y. Yamaguchi, H. Iwasaki, N. Kobayashi, and T. Komatsubara, Physica B **177**, 169 (1992).
[11] J. Rossat-Mignod, in *Methods of Experimental Physics 23, Part C*, edited by K. Sköld and D. L. Price (Academic, Orlando, 1987), p. 131.
[12] W. Bazela, J. Leciejewicz, and A. Szytula, J. Magn. Magn. Mater. **50**, 19 (1985).
[13] A. Szytula, W. Bazela, and J. Leciejewicz, J. Phys. (Paris) Colloq. **49**, C8-383 (1988).
[14] S. Quezel, J. Rossat-Mignod, B. Chevalier, W. X. Zhong, and J. Étourneau, C. R. Acad. Sci. Ser. II **301**, 919 (1985).