

Multi- k magnetic structures in $\text{USb}_{0.9}\text{Te}_{0.1}$ and $\text{UAs}_{0.8}\text{Se}_{0.2}$ observed via resonant x-ray scattering at the $U M_4$ edge

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Experiments with resonant photons at the $U M_4$ edge have been performed on a sample of $\text{USb}_{0.9}\text{Te}_{0.1}$, which has an incommensurate magnetic structure with $k=k=0.596(2)$ reciprocal lattice units. The reflections of the form $\langle kkk \rangle$, as observed previously in a commensurate $k=1/2$ system [N. Bernhoeft *et al.*, Phys. Rev. B **69**, 174415 (2004)], are observed, removing any doubt that these occur because of multiple scattering or high-order contamination of the incident photon beam. They are clearly connected with the presence of a $3k$ configuration. Measurements of the $\langle kkk \rangle$ reflections from the sample $\text{UAs}_{0.8}\text{Se}_{0.2}$ in a magnetic field show that the transition at $T^* \sim 50$ K is between a low-temperature $2k$ and high-temperature $3k$ state and that this transition is sensitive to an applied magnetic field. These experiments stress the need for quantitative theory to explain the intensities of these $\langle kkk \rangle$ reflections.

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I. INTRODUCTION

In determining magnetic structures for a vast array of materials, neutron diffraction is the technique of choice and has been used with great success for the last 50 years. However, the realization by Kouvel and Kasper¹ that multi- k configurations, in their case in $(\text{Ni},\text{Fe})_3\text{Mn}$ alloys, could be present, often makes the ground-state determination by neutron diffraction ambiguous. Single- k configurations have domains with a single propagation direction within a domain, whereas in the multi- k configurations several propagation directions, commonly 3 in cubic systems but possibly more, exist *simultaneously*.^{2,3} The common method to examine whether configurations are single- or multi- k is to apply an external perturbation (usually magnetic-field cooling) and to measure the intensities from various “domains.” If they are changed by the perturbation, then the configuration can often be suggested,^{2,4} whereas if they are not, a multi- k configuration is assumed. Unfortunately, one can also envisage that the field cooling itself changes the ground state of the system or that the field used is not large enough to change the domain population. Thus, the confusion of whether a magnetic structure is multi- k or not continues to be a limitation of neutron diffraction studies of magnetic materials.

With this background, it was therefore of considerable interest when additional diffraction peaks observed in resonant x-ray scattering (RXS), apparently associated uniquely with the $3k$ configuration, were reported for a sample of $\text{UAs}_{0.8}\text{Se}_{0.2}$.⁵

These diffraction peaks were weak (less than 10^{-4} of the main Bragg peaks arising from magnetic order) but clearly observable in the RXS experiments because of the large resonant enhancement that occurs when the incident photon energy is tuned to the M_4 edge of uranium.⁶ More recently, these extra reflections have been observed also using neutron diffraction,⁷ confirming that the reflections are indeed magnetic dipole in origin. In addition, the neutrons demonstrate

that the effect is not connected with the surface, which is always a possibility if effects are observed only with the RXS technique.

The new reflections are associated with the phase coherence of the three different propagation directions that make up the $3k$ configuration.⁸ Such reflections were reported in the assumed $3k$ structure of CeAl_2 .⁹ In this latter paper, the term giving rise to the $\langle kkk \rangle$ reflections, which is a convenient shorthand notation for this subset of reflections since they contain a contribution from each k component, was identified with a fourth-order term in the free-energy expansion. The term is proportional to $M_x M_y M_z M_c$, where the individual components of the magnetization propagating along the three perpendicular axes are M_x , M_y , and M_z and the term $k_c = k_x + k_y + k_z$ represents the *coherent* part of the different magnetization distributions. Note that expressing such terms within free energy fulfills the symmetry of the system, but gives no idea of their intensity with respect to the main $\langle k00 \rangle$ magnetic Bragg diffraction peaks, and we have not found any published material predicting the possible intensities of such reflections. However, further research on CeAl_2 established that these reflections came from a different magnetic configuration in the system,¹⁰ and our present understanding of CeAl_2 is that it is *not* a $3k$ system,^{11,12} so these reflections should not be present. Despite doubts of the validity of the experimental results in Ref. 9, the theoretical expression in this reference still gives the correct symmetry considerations for the $\langle kkk \rangle$ reflections.

The present paper brings further experimental evidence about these unusual diffraction peaks. So far, studies of the $\langle kkk \rangle$ peaks^{5,7} have been confined to samples with the commensurate magnetic wave vector $k=1/2$. Although significant efforts were made in Refs. 5 and 7 to eliminate multiple scattering and higher-order diffraction effects, one way to ensure that such effects are further minimized is to examine a material in which the magnetic ordering is *incommensurate*. By exploiting the magnetic properties found in the sys-

tem USb-UTe,¹³ the first part of this paper presents clear evidence for the different diffraction peaks in such an incommensurate magnetic system.

In the second part of the paper, we return to the commensurate system UAs_{0.8}Se_{0.2} with $k=1/2$ and examine the behavior of the $\langle kkk \rangle$ reflections as a function of magnetic field. Bulk techniques (specific heat and magnetic susceptibility) performed on a sample of the same composition have shown anomalies associated with what is believed to be the transition between a $2k$ state at low temperature and a $3k$ state at higher temperature,^{5,14} and our goal was to study the field dependence of the different diffraction peaks to determine the exact nature of the transition. The experiments show that the transition as a function of applied field is, indeed, one between the $2k$ and $3k$ states.

II. EXPERIMENTAL DETAILS AND DISCUSSION

All RXS experiments reported here have been performed on the ID20 beamline at the European Synchrotron Radiation Facility, Grenoble, France,¹⁵ using Si(111) horizontally focusing double crystal monochromator. Two Si-coated mirrors assure vertical focusing at the sample position and elimination of higher harmonics from the incident beam. Au(111) single crystal was chosen as a polarization analyzer based on the Bragg diffraction close to the Brewster angle of $\theta=45^\circ$ at the energy of U M_4 edge $E=3.724$ keV.

Both compounds studied in these experiments have the cubic rocksalt crystal structure, space group $Fm\bar{3}m$, No. 225 with lattice parameters $a=6.203$ Å for USb_{0.9}Te_{0.1} and $a=5.773$ Å for UAs_{0.8}Se_{0.2}. The doping anions Te and Se substitute randomly on the anion sublattice of the NaCl crystal structure and simply act to tune the electron configuration, see Refs. 2 and 4. The magnetic structures are longitudinal and described by the propagation vector k . Diagrams, and more details, of the multi- k arrangements are given in Fig. 1 of Ref. 5.

In the case of USb_{0.9}Te_{0.1}, the sample was installed in a closed-cycle cryostat with base temperature of about 10 K, in the vertical scattering geometry. This setup, with [100] direction vertical, allows for partial rotation of the sample around the scattering vector, necessary for azimuthal scans. The UAs_{0.8}Se_{0.2} experiment was performed in the horizontal scattering geometry, with the sample mounted in a 10 T cryomagnet. The magnetic field was applied along the $[1\bar{1}0]$ direction, allowing reflections within the (hhl) plane to be measured. We used polarization analysis of the scattered beam only in the first part of the experiment to confirm that our signal was uniquely in the rotated polarization channel ($\pi \rightarrow \sigma$, where π and σ are the beam polarization components perpendicular and parallel to the scattering plane). We know from earlier work⁵ that these $\langle kkk \rangle$ reflections are magnetic dipolelike in their response. Thus, the use of polarization analysis assures that the signal in the $\sigma \rightarrow \pi$ channel contains no contribution from any lattice effects (that would have a $\sigma\sigma$ contribution) and is magnetic only. The main part of the experiment was performed without analysis of the scattered beam polarization for intensity reasons because the

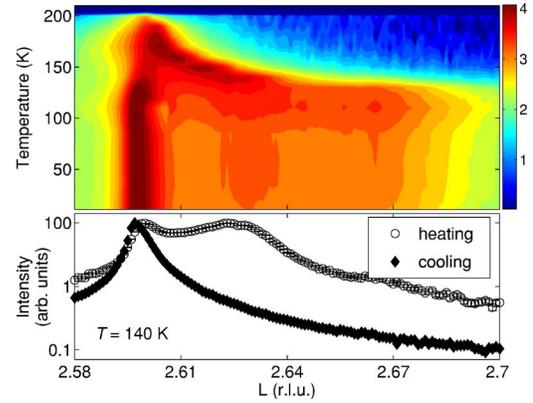


FIG. 1. (Color online) Upper panel: Magnetic intensity from the sample of USb_{0.9}Te_{0.1} as a function of scattering vector (00L) and temperature. The sample was first cooled to 10 K and then data were collected as the sample was heated. Lower panel: Data at $T=140$ K on both heating and cooling. The main magnetic modulation corresponds to $k \sim 0.6$ rlu, originating from the (002) charge reflection, but intensity is observed over a wide range of wave vector. At the highest temperatures, just below $T_N \sim 200$ K, a single incommensurate magnetic wave vector is found, in agreement with the phase diagram proposed in Ref. 13. The scale in both panels is logarithmic.

$\langle kkk \rangle$ peak intensity is further reduced due to the poor transmission through the Be windows of the cryomagnet. Fortunately, for the scattering vector of interest, $Q=(1/2 \ 1/2 \ 5/2)$, the scattering angle 2θ is close to 97° , which means that with incident beam π polarization, any background due to a lattice contribution (like multiple scattering), corresponding to a $\pi\pi$ signal, is negligible.

The specific heat was measured on an 83 mg sample using the relaxation method in a PPMS-9 equipment (Quantum Design) within the temperature range of 2–300 K. The external magnetic field up to 9 T was applied along the [001] direction.

A. Experiments on USb_{0.9}Te_{0.1}

The magnetic phase diagram of the USb-UTe solid solutions was first examined 25 years ago.¹³ USb is a well-known (cubic) $3k$ antiferromagnet with wave vector $\langle k00 \rangle$, where $k=1$ and $T_N=215$ K. With a small doping of Te, the magnetic wave vector (k) starts to reduce and, at least near T_N , incommensurate magnetic order is found.¹³ For our experiments, we have used a large crystal (because neutron experiments were also performed on this sample) of 0.512 g, which has a good mosaic of 0.026° full width at half maximum.

An overview of the magnetic scattering as a function of temperature is given in Fig. 1. The scattering arises from magnetic modulations with wave vectors (k) extending from ~ 0.6 to ~ 0.7 reciprocal lattice units (rlu). The pattern shows a more complicated situation than reported earlier.¹³ Presumably, this may be explained by the poorer resolution of the early neutron experiments as well as the much larger intensity, and thus sensitivity, obtained in the RXS technique. All

intensities in this plot are measured in the $\sigma \rightarrow \pi$ channel, and since there is no scattering in the $\sigma \rightarrow \sigma$ channel, the scattering is magnetic in origin. At high temperatures, near T_N , the scattering is incommensurate and consists of one modulation with ~ 0.6 rlu, as reported,¹³ but below about 160 K, the material exhibits frustration with magnetic scattering distributed over a wide range of modulation vectors. In addition to this extension of the modulation along the longitudinal direction $[00L]$ (as shown in Fig. 1), the scattering in the transverse direction is broad in q as well, showing the short-range ordering of the magnetic correlations. Figure 1 shows that at all temperatures there is a strong and relatively sharp component at $k=0.6$, and this, rather than a modulation at $k=2/3=0.667$ rlu, is clearly the dominant correlation in the system. Moreover, by slow cooling of the sample, it was found that the $k \sim 0.6$ rlu modulation was maintained to at least 140 K. An accurate measure of $k = 0.596(2)$ rlu was made at this temperature. All subsequent experiments were performed at 140 K.

On cooling into the ordered state, no sign of any *external* structural distortion (i.e., a distortion from cubic to tetragonal) was observed. This is consistent with the fact that all the antiferromagnet configurations in the USb-UTe solid solutions are $3k$ in nature.¹³

In agreement with previous work on the UAs-USE solid solutions, we found the peaks referred to as $\langle k k 0 \rangle$.¹⁶ These arise from the E1-F^[2] term in the RXS cross section¹⁷ and are commonly associated with the quadrupole interactions in these materials. They are characterized by a sharp photon energy dependence of the scattering, intensity in both $\sigma \rightarrow \pi$ and $\sigma \rightarrow \sigma$ channels, and a characteristic azimuthal dependence of the scattered intensity.¹⁶

The reflections characterizing the $3k$ magnetic structure have the form $\langle k k k \rangle$,⁵ and we show two such reflections in Fig. 2. Comparison of the intensities between data in Fig. 2 gives approximately a factor of $(3 \times 10^{-5}/0.6) \times (0.0060/0.0032)^2 = 1.7 \times 10^{-4}$, where the first term is simply the intensity of the peak and the second factor takes into account the different widths in the H and L directions (the resolution in the K direction of the spectrometer is poor and integrates all the signal). This is consistent with previous estimates.⁵ It is noteworthy that the correlations between the different components in the $3k$ structure give rise to a correlation length that is almost half that of a single k component. This was not previously observed⁵ because for a *commensurate* system (like in the UAs-USE) the strain terms locking the magnetic and charge modulations in the free energy ensure that the magnetic correlations are as long range as the charge (see Fig. 2 of Ref. 5). However, this is not the case in an incommensurate system as examined here.

As reported earlier,⁵ all the signals for the $\langle k k k \rangle$ peaks was in the $\sigma \rightarrow \pi$ channel and the photon energy dependence was similar to that of the $\langle k 0 0 \rangle$ peak, i.e., a Lorentzian centered at the U M_4 resonance. This assures that there is no charge contribution to these reflections.

An azimuthal scan of the same reflection is shown in Fig. 3. The dipole component from which this reflection arises is $[-111]$, as shown by the simulation of the azimuthal dependence from such a component, scaled to the maximum ob-

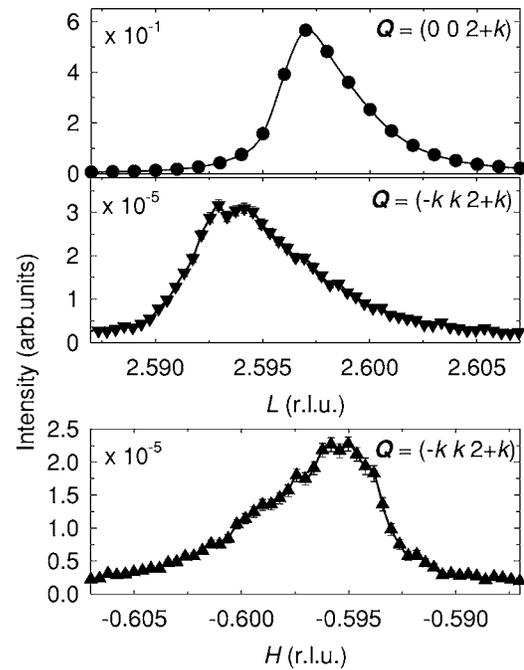


FIG. 2. Comparison of intensity profiles of a $\langle k 0 0 \rangle$ peak (upper panel, as in Fig. 1) and $\langle k k k \rangle$ peak (lower two panels) at $T = 140$ K. Notice that the asymmetry extends to higher k value on both $\langle k 0 0 \rangle$ and $\langle k k k \rangle$ peaks and in both L scan and H scan. A small difference in the actual value of k for $\langle k 0 0 \rangle$ and $\langle k k k \rangle$ peaks is due to crystal and/or diffractometer misalignment. The true value of k is determined from the positions of the third harmonics reflections $(0\ 0\ 3k)$ and $(0\ 0\ 4-3k)$ to be $0.596(2)$ rlu. The $\langle k 0 0 \rangle$ peak has a full width at half maximum (FWHM) of 0.0036 rlu in the L direction and 0.0032 rlu in the transverse (H) direction. This may be compared to 0.0023 and 0.0014 rlu, respectively, for the charge peaks measured at the same photon energy. The latter are close to the instrumental resolution, which is poor in the χ direction (K). The $\langle k k k \rangle$ peaks have FWHM's of about 0.0060 rlu in both the L and H directions.

served value and with no other variable. It should be realized that the azimuthal dependence of the $\langle k 0 0 \rangle$ reflections corresponds to a dipole moment along a $\langle 100 \rangle$ direction; thus the $\langle k k k \rangle$ reflections, which have their dipole moment corresponding to one of the $\langle 111 \rangle$ directions, show the essential $3k$ nature of the configuration. This is discussed in Ref. 7, where

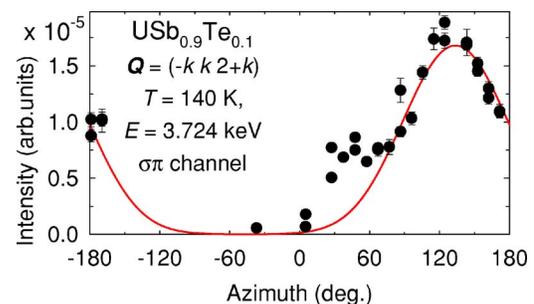


FIG. 3. (Color online) Azimuthal scan of the $(-k k 2+k)$ reflection. The solid line is a fit to model in which the dipole moment for the reflection rotates around the direction $[-111]$.

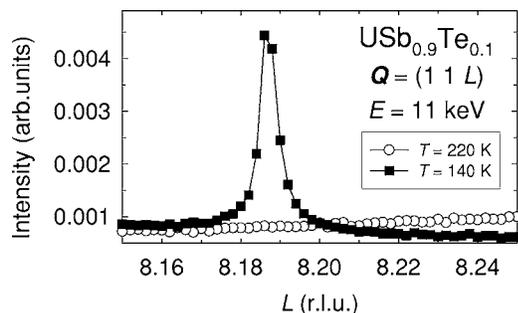


FIG. 4. Scan along $[11L]$ direction with the photon energy at 11 keV and a Ge(111) analyzer installed in the PA stage. The very sharp peak at $(1\ 1\ 7+2k)$ corresponds to the magnetoelastic term. It gives an excellent measure of the magnitude of k .

a similar azimuthal dependence was found in the $\text{USb}_{0.85}\text{Te}_{0.15}$ material.

Finally, as pointed out first by McWhan *et al.*,⁶ and later in more detail in the UAs-USE system by Longfield *et al.*,¹⁸ there should be a magnetoelastic distortion in these systems corresponding to peaks at $\langle 2k\ 0\ 0 \rangle$. This is a charge term corresponding to the charge-density wave riding on the underlying magnetic modulation. The intensity of this peak increases as Q^2 , where Q is the scattering vector. Since the displacement-wave amplitude is small, both the penetration and limited Q range available at 3.7 keV mitigate against the observation of such peaks. Thus, the photon energy was increased to 11 keV and a Ge(111) crystal mounted in the polarization stage. A resultant scan along the direction $[11L]$ is shown in Fig. 4. This clearly shows the magnetoelastic $2k$ component and, as expected, that it exists in an incommensurate system.

In summary, for the $\text{USb}_{0.9}\text{Te}_{0.1}$ we emphasize that the magnetic modulation is incommensurate, but that the $\langle kkk \rangle$ peak exists at the same level of intensity ($\sim 10^{-4}$) as compared to the standard $\langle k00 \rangle$ magnetic reflections. These peaks, as seen by x rays, arise from the phase coherence between the different components in the $3k$ structure and are not artifacts of the data collection or reflections arising from higher-order components in the photon beam.

B. Experiments on $\text{UAs}_{0.8}\text{Se}_{0.2}$

In the previous work on this system,^{5,7} it was shown that the $\langle kkk \rangle$ reflections appeared to disappear (on cooling) below T^* (~ 50 K) and thus corresponds to a transition between the high-temperature $3k$ to the lower-temperature $2k$ state, in which state the reflections should not exist, as originally proposed by Kuznietz *et al.*¹⁴ Bulk measurements of specific heat and magnetization showed anomalies also at this temperature.⁵ An overview of the specific-heat data with the original sample (I) is shown in Fig. 5.

With synchrotron experiments on a new sample of $\text{UAs}_{0.8}\text{Se}_{0.2}$ (sample II), we have first reexamined the nature of the transition at T^* . Figure 6 shows (upper panel) the temperature dependence of a $\langle kkk \rangle$ reflection and (lower panel) the behavior of the lattice as measured with high-resolution photons. Below T^* , there is an external lattice dis-

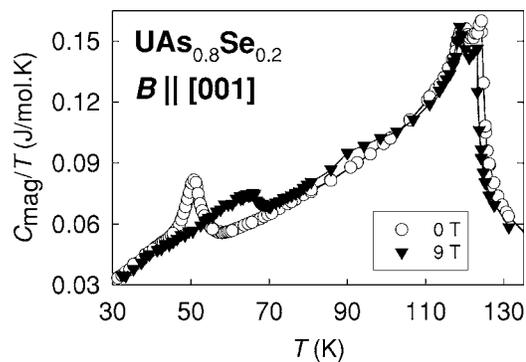


FIG. 5. Specific heat measured on sample I of $\text{UAs}_{0.8}\text{Se}_{0.2}$ as a function of magnetic field. The ordering at $T_N \sim 125$ K is insensitive to magnetic field, but the transition (T^*) at ~ 50 K increases with increasing B .

tortion to at least a tetragonal symmetry (and may be lower), and this results in a splitting of the (008) charge reflection. These results are consistent with the idea that the material transforms on cooling from the high-temperature $3k$ state, which has cubic symmetry, to the low-temperature $2k$ state, which has lower than cubic symmetry.

To compare the data taken with specific heat and RXS, we have chosen to normalize the data to T^* as the specific heat were taken on sample I and RXS measurements on sample

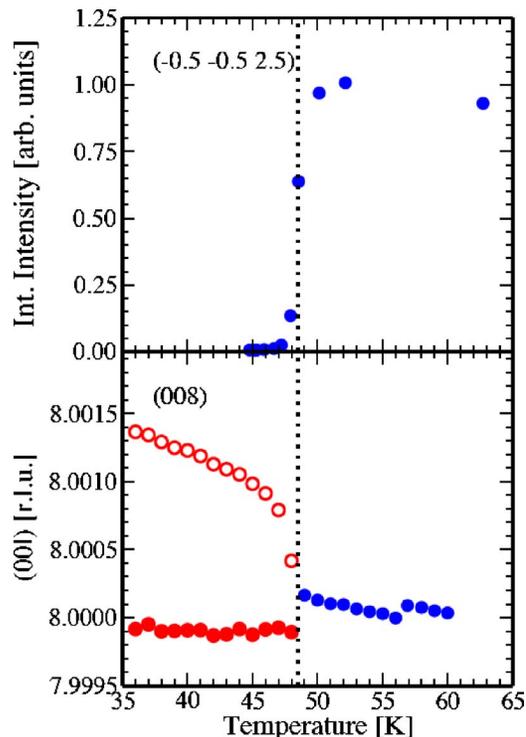


FIG. 6. (Color online) Upper panel: Intensity of the $(-0.5\ 0.5\ 2.5)$ reflection from sample II on heating from low temperature and with a photon energy tuned to the U M_4 resonance. Lower panel: Position of the lattice peak as measured with high-resolution 8 keV photons+Ge(111) analyzer. A double peak is observed below T^* , indicating an external lattice distortion to tetragonal (or lower) symmetry.

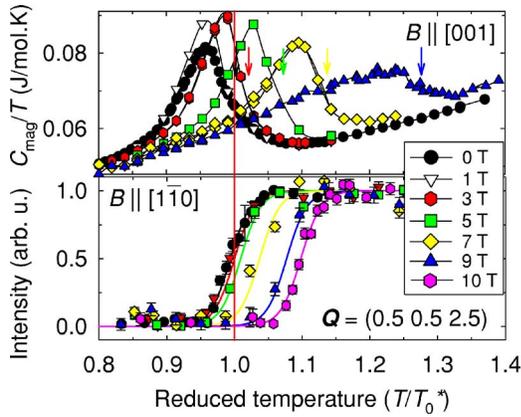


FIG. 7. (Color online) Specific heat (upper part) on sample I divided by T and with the phonon contribution subtracted and (lower part) intensity of the $(0.5\ 0.5\ 2.5)$ reflection in sample II of $\text{UAs}_{0.8}\text{Se}_{0.2}$ as a function of temperature normalized to T^* at $B=0$. The arrows in the upper panel indicate T^* . Notice that the magnetic fields have been applied in two different directions for the different techniques.

II. Small differences in Se composition may account for a small difference in T^* of about 3 K; compare Figs. 5 and 6. Furthermore, the original specific-heat data were taken with $B\parallel[001]$, whereas the RXS measurements with $B\parallel[1\bar{1}0]$. The latter geometry is required to obtain the $\langle kkk \rangle$ peak in the equatorial plane of the magnet. However, the point of this study is to show that the change in the intensity of the $\langle kkk \rangle$ reflection as a function of applied field follows (approximately) the phase transition, and thus identifying it as one between $2k$ ($T < T^*$) and $3k$ ($T > T^*$), rather than understanding the full (B, T) phase diagram of this material.

Figure 7 (upper part) shows the change of specific heat (divided by T and with the phonon contribution subtracted) and the intensity of a $\langle kkk \rangle$ reflection as a function of $T/T_{B=0}^*$ for different applied magnetic fields. In addition to the fact that both measurements show T^* rising as a function of applied field, there are two further points of note in this figure.

The first is that the rise of T^* is much greater for $B\parallel[001]$ than $B\parallel[1\bar{1}0]$. Since the phase diagram as a function of applied field has not been reported for $\text{UAs}_{0.8}\text{Se}_{0.2}$, our knowledge is necessarily limited. However, studies of UAs (Refs. 2, 4, and 21) and $\text{UAs}_{0.75}\text{Se}_{0.25}$ (Ref. 20) give some indication of what might occur. In both cases, the easy axis is not $\langle 100 \rangle$, but the initial transitions occur at lower fields (at constant temperature) with $B\parallel[001]$ than any other direction. So the transition at T^* is more rapidly shifted with $B\parallel[001]$ than $B\parallel[1\bar{1}0]$, as observed in Fig. 7. Further details of these effects as a function of magnetic-field direction may be found in Ref. 21.

The second observation is related to the surprising result for the specific heat for $B=9$ T, which shows a broad peak extending over many degrees. In contrast, the breadth of the transition, over which the $\langle kkk \rangle$ reflection develops for high fields, is about the same as at lower fields. Although, as in the first part, we have not done sufficient experiments to verify the exact nature of the high-field state, it seems likely

that a second transition occurs for fields near 9 T and above. Again, taking the work on UAs (Refs. 2, 4, 19, and 21) and $\text{UAs}_{0.75}\text{Se}_{0.25}$ (Ref. 20) as a guide, a transition to a modified magnetic structure occurs for these high fields. With $B\parallel[001]$ the transition involves a change in the modulation component parallel to B ,¹⁹ but this may not be relevant with $B\parallel[1\bar{1}0]$. Perhaps the surprising result is not that the specific-heat anomaly at these high fields is so smeared over many degrees, but that the intensity of the $\langle kkk \rangle$ reflection shows clearly that whatever the magnetic state is above T^* , it is certainly $3k$ in nature.

These results shown in Fig. 7 leave little doubt that the transition at T^* is between the $2k$ (for $T < T^*$) and $3k$ (for $T > T^*$) states, with the $2k$ state being stabilized with increasing magnetic field.

III. CONCLUSIONS

Although the earlier papers in this series^{5,7} provided strong evidence that the different $\langle kkk \rangle$ peaks were associated with the $3k$ nature of the magnetic configuration, some doubts remained. In particular, the observation of very weak reflections at highly symmetric lattice points must always be regarded with caution as complicated effects from higher-order components in the incident photon or neutron beam or multiple scattering may always produce weak reflections at these special positions. We are dealing with intensities of the order of 10^{-4} of the magnetic peaks, which, in the case of neutrons or RXS at the U M edges, represent an intensity of $\sim 10^{-6}$ of the nuclear or charge peaks, respectively. The present experiments on a $3k$ incommensurate system ($\text{USb}_{0.9}\text{Te}_{0.1}$) show unambiguously that the $\langle kkk \rangle$ reflections exist and behave in the same way as previously reported.⁵ They clearly arise from the coherence of the individual three components in the system.

Furthermore, a careful examination of the transition at T^* in $\text{UAs}_{0.8}\text{Se}_{0.2}$, which had been proposed¹⁴ as a transition between the $2k$ (low temperature) and $3k$ (high temperature) states, shows that the field dependence of the $\langle kkk \rangle$ reflection confirms this proposal. As the field is increased, the $2k$ state is stabilized at the expense of the $3k$ modulations.

The observation and explanation of these $\langle kkk \rangle$ reflections in terms of the free energy, as proposed⁹ in connection with CeAl_2 , are satisfying from a conceptual point of view as to what symmetry considerations define these $\langle kkk \rangle$ peaks. An alternative analysis in terms of Clifford algebra for the presence of these peaks has also been recently advanced.²² In neither case do the arguments specify the intensity to be expected. Other experiments that we are undertaking are measuring the form factor $f(Q)$ of the $\langle kkk \rangle$ reflections with neutron scattering so as to determine the spatial distribution of the magnetization that is involved in the coherence between the different $3k$ components.

More than 40 years after the discovery of multi- k magnetic structures,¹ some aspects of their nature remain elusive; it is surprising, for example, that $\langle kkk \rangle$ reflections have not been observed previously. Ironically, multi- k configurations have been predicted²³ to be the stable magnetic states in

nanoantiferromagnets, simply because of the high cost in energy of domain walls. Perhaps this will give a further motivation for fully understanding the subtleties of multi- k structures.

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