X-Ray Magnetic Scattering in Antiferromagnetic URu_2Si_2

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X-ray-resonance magnetic scattering has been used to study antiferromagnetic ordering in the small-moment ($\bar{\mu} \cong 0.02\mu_B$) heavy-fermion superconductor URu_2Si_2. The intensity of the magnetic (003) reflection develops abruptly at $T_N \simeq 17$ K and grows linearly to $T = 3$ K, where it saturates. Long-range antiferromagnetic order ($\zeta^c_{003} \cong 450$ Å) persists into the superconducting state at $T_c = 1.3$ K demonstrating the microscopic coexistence of these two ground states. At saturation, a remarkable peak intensity of 8 counts/sec was measured.

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The low-temperature properties of heavy-fermion superconductors, which arise from the quantum-mechanical contact between the local f electrons and the metallic d band,[1] continue to reveal a rich variety of magnetic and superconducting ground states. Of particular interest are URu_2Si_2,[2] UPt_3,[3] and UBe_{13},[4] where both antiferromagnetism and superconductivity are present. In these systems there is the possibility that spin fluctuations provide the pairing mechanism. Neutron-scattering studies in URu_2Si_2 have shown that a weak antiferromagnetic state which develops at low temperatures persists into the superconducting state below $T_c = 1.2$ K.[2] However, a certainty of a microscopic coexistence of these two states in URu_2Si_2 is somewhat clouded by the relatively short magnetic correlation lengths measured in that experiment. On the other hand, in UPt_3 it has been demonstrated that static magnetic order plays an integral role in the different superconducting states of that system.[3]

We have studied the static antiferromagnetic order in a high-quality, single crystal of URu_2Si_2 to $T = 0.9$ K with x-ray magnetic scattering. The intensity of the (003) magnetic Bragg reflection develops abruptly at $T_N \simeq 17$ K, where the large λ anomaly in the specific heat has been observed,[5] unlike the broad onset of previous neutron studies.[2] The (003) intensity grows linearly over an unusually broad temperature range down to $T \simeq 3$ K $\simeq T_N/6$, where it saturates and subsequently remains constant, within the error bars of this experiment, into the superconducting phase below $T_c = 1.3$ K. We estimate the ordered moment to be $(0.02 \pm 0.01)\mu_B$ at saturation, where a remarkable peak intensity of 8 counts/sec was measured. In addition, we have measured magnetic correlation lengths, which are approximately independent of temperature below T_N and are $\zeta^c_{003} \cong 450$ Å along the c axis and $\zeta^{a\text{-}b}_{003} \cong 200$ Å in the basal plane. These results and the agreement with a recent neutron-scattering study on the same sample[6] demonstrate that x-ray magnetic scattering is a useful new probe to study antiferromagnetic order in small-moment systems.

These new results have been made possible for two reasons: (i) The URu_2Si_2 sample employed in these experiments was of a better quality (higher T_c with fewer stacking faults) than the one employed in previous neutron studies and (ii) the naturally higher q resolution of the synchrotron x-ray beam relative to a typical neutron study has allowed us to probe larger correlation lengths.

The ability to measure as small a moment as in URu_2Si_2 has been made possible by the recent discovery of x-ray-resonance magnetic scattering. When the energy of the incident synchrotron x-ray beam was tuned through the U M_{IV} absorption edge ($E = 3.728$ keV) in UAs an enhancement of the magnetic scattering intensity of 10^7 (Ref. 7) was observed relative to the intensity far above the M_{IV} edge at $E = 7.6$ keV.[8] The resonance process, which is described in detail elsewhere,[9] arises from electric multipole transitions between the polarized (magnetic) ground state of the atom and an intermediate bound state. This bound state consists of a core hole and the Fermi edge states, and can be thought of as an $N+1$ impurity state, where N is the atomic number. Through atomic selection rules associated with these multipole transitions and the polarization of the incident x-ray beam, this resonant process is sensitive to the axis of quantization of the atom, and thus, antiferromagnetic order. The M_{IV} edge in U corresponds to a dipole transition and involves the coupling of the $3d_{3/2}$ core hole to the unoccupied $5f_{5/2}$ state near the Fermi edge. This is the same $5f_{5/2}$ state that gives rise to the various ground states in URu_2Si_2. The details of the unoccupied $5f_{5/2}$ Fermi edge structure have recently been explored with x-ray-resonance Raman scattering with 1-eV resolution and will be described elsewhere.[10]

The x-ray magnetic scattering (XRMS) experiments were carried out on the Oak Ridge National Laboratory beam line X14 at the National Synchrotron Light Source. The salient features of X14 are described in our previous XRMS paper.[7] The URu_2Si_2 sample was mounted on a copper cold finger on the central axis of a ^{3}He refrigerator, centered in a four-circle diffractometer in the vertical scattering plane geometry with x-ray access to the sample provided by a $\frac{1}{4}$-mm-thick cylindrical Be window in the Dewar vacuum can and two successive 6-μm-thick aluminized Mylar windows in the heat shields at 77 and 4 K, respectively. A mutual-inductance coil, used for monitoring the ac susceptibility, was placed around the sample but out of the x-ray beam path. A 10% change in signal, which consisted of the sample plus a large background, was observed at T_c. Finally, a Si(Li) solid-state detector was employed to detect the magnetic reflections.

For dipole transitions in XRMS the polarization of the scattered photon is rotated by 90° (π polarization) relative to a linearly polarized incident photon (σ polarization).[9] Since the polarization of the charge scattering is unrotated, a factor-of-70 improvement in the magnetic signal compared to background (mostly diffuse charge scattering) was achieved with a polarization analyzer. A LiF(111) crystal, with a 10% peak reflectivity, was chosen as an analyzer since at 3.724 keV the Bragg angle (θ) is approximately 45°.[11] The analyzer crystal, with a mosaic spread of 0.1°, defined the momentum resolution transverse to the vertical scattering plane to be $\Delta q_x/q \sim 4\times 10^{-3}$. In the scattering plane the momentum resolution was approximately $\Delta q_z/q \sim 2\times 10^{-3}$ and $\Delta q_y/q \sim 10^{-3}$, defined by the vertical divergence of the synchrotron beam and the mosaic of the URu_2Si_2 crystal, respectively. In previous neutron-scattering experiments the corresponding resolutions were $\Delta q_x/q \sim 0.06$ and $\Delta q_z/q \sim 0.06$.[2]

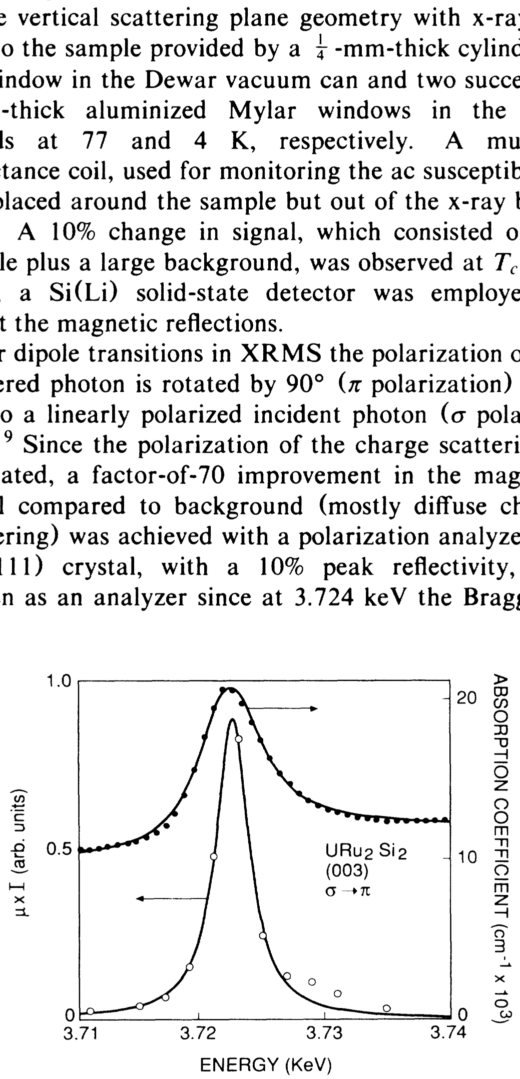

FIG. 1. The open circles are the peak intensity of the (003) magnetic Bragg reflection vs energy. The polarization of the incident synchrotron radiation is linear and perpendicular to the vertical scattering plane (σ), and upon scattering the polarization is rotated 90° into the vertical scattering plane (π). The solid line through the open circles is a fit with a Lorentzian line shape whose width is approximately 5 eV. The solid circles are the absorption coefficient vs energy, derived from a fluorescence spectrum.

The diffraction experiments were carried out on a 3-mm-thick URu_2Si_2 single crystal cut from a cylindrical boule with a c-axis face. URu_2Si_2 has the $ThCr_2Si_2$ structure with the U atoms on a body-centered tetragonal lattice. The positions of the Bragg peaks are expressed in terms of reciprocal-lattice units $a^* = b^* = 2\pi/a = 1.524$ Å^{-1} and $c^* = 2\pi/c = 0.656$ Å^{-1}. The measurements were carried out with momentum transfer along [001]. URu_2Si_2 exhibits type-I antiferromagnetic order corresponding to a doubling of the unit cell along the c axis with the moments parallel to the c axis. The Néel temperature, which has been associated with the large λ anomaly in the specific heat, is $T_N \cong 17$ K.

Figure 1 shows the energy dependence of the peak intensity of the (003) magnetic reflection corrected for absorption. At the peak of the resonance we collected a remarkable 8 counts/sec, which is comparable with recent neutron measurements carried out at the Chalk River Nuclear Laboratories reactor, where 2 counts/sec was measured.[12] The peak in the resonance profile is at approximately the same energy as the peak in the absorption spectrum, which is shown at the top of Fig. 1, and is similar to what has been observed at the M_{IV} edge in UAs (Refs. 7 and 8) and UN.[13] The magnetic resonance process arises from the excitonic feature seen as the so-called white line in the absorption spectrum. The penetration depth at the peak of the resonance was approximately 5000 Å. Note that these data, as well as all subsequent data described below, reflect only the rotated component of polarization of the scattered beam. Without the resonance enhancement,[7] x-ray magnetic scattering from a moment of $0.02\mu_B$ would have been too weak to observe.

The integrated intensity of the (003) magnetic reflection, measured with the incident x-ray energy set at the peak in the resonance profile in Fig. 1, is shown in Fig. 2. The intensity grows linearly over an unusually broad temperature range, from its onset at $T = 17$ to 3 K where it saturates. The abruptness of the saturation is somewhat uncertain due to statistics and systematic uncertainties. The saturation moment was estimated by comparing the intensity at the (003) magnetic reflection, normalized to the intensity of the (002) charge reflection, to the normalized resonance magnetic intensity in UAs (see Ref. 7). We found from Fig. 2 that $\bar{\mu} \cong (0.02 \pm 0.01)\mu_B$. The discrepancy between our re-

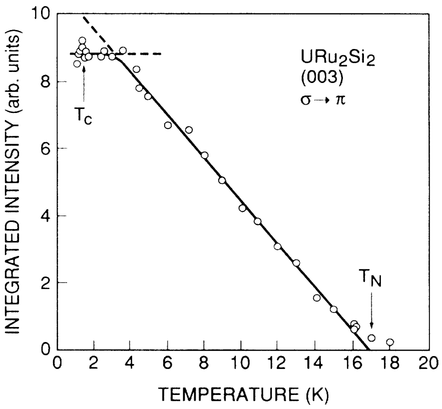

FIG. 2. Integrated intensity of the (003) magnetic Bragg reflection vs temperature. Notice the unusually broad linear regime, from T_N to $T_N/6$. The solid line is a guide to the eye. No change in the ordered moment is observed at the superconducting temperature T_c. Saturation corresponds to 8 counts/sec at the Bragg peak.

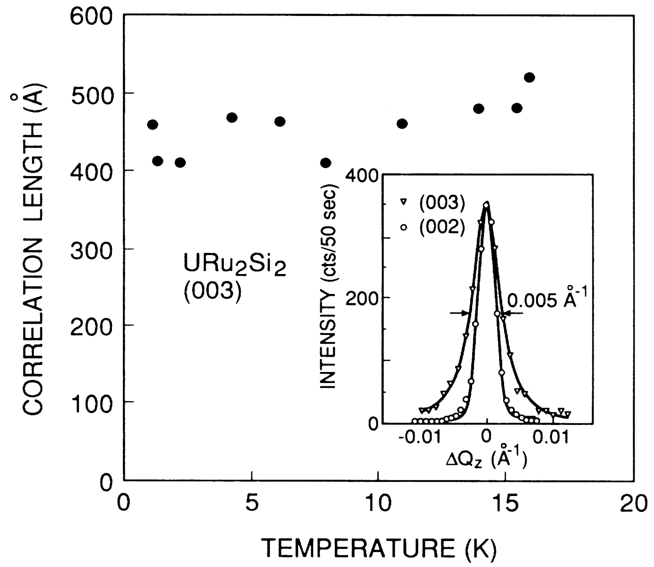

FIG. 3. Correlation length vs temperature along the c axis at the (003) magnetic Bragg reflection. Inset: Radial scans through the (003) magnetic reflection and the (002) charge reflection. The width of the (002) reflection puts an upper limit on the resolution width at the (003).

sult and the neutron-diffraction determinations[2,6,12] is probably due to the error incurred upon normalization to a separate experiment (UAs). No change in the ordered moment was observed at or below the superconducting transition at $T_c \cong 1.3$ K to within the error bars ($\pm 10\%$) of this experiment. The scatter in the low-temperature data points resulted from the systematic uncertainty associated with mechanical stability of the crystal with respect to the diffractometer while pumping on both the ^{4}He and ^{3}He. Our results confirm previous neutron observations of the order parameter that show saturation occurs at 2.5 K, above the superconducting transition.[14]

In addition, we have measured the correlation length of the antiferromagnetic order along the tetragonal c axis as well as in the basal a-b plane. The scans were not resolution limited as can be seen from the inset in Fig. 3. The longitudinal (c-axis) spectrometer resolution width was estimated by measuring the longitudinal width at the (002) charge peak, which was then removed from the (003) width. This approximation gives a lower limit for the magnetic inverse correlation length because the (003) reflection is nearly dispersionless, while the (002) is not and, therefore, is somewhat broadened (see, for example, Ref. 15). The correlation length was defined as the inverse of the linewidth, $\zeta \equiv 1/\kappa$, and is shown as a function of temperature in Fig. 3. Along the tetragonal c axis ζ_{003}^{c} is approximately constant with a value of 450 Å. The correlation length in the basal plane, $\zeta_{003}^{a\text{-}b}$, is approximately 200 Å. The smaller correlation length of 200 Å along the c axis measured previously with neutrons was probably due to poorer sample quality, i.e., stacking faults as suggested by Ref. 2. Although the magnetic correlations that we have measured are anisotropic, they extend an equal number of interplanar spacings, i.e., $\zeta_{003}^{c}/\zeta_{003}^{a\text{-}b} \sim c/a$. Finally, we note that no magnetic scattering in the critical region just above T_N was observed.

An unusual feature of the temperature dependence of the magnetic intensity is the broad range of linear growth below $T = T_N = 17$ K. Inelastic neutron studies in URu$_2$Si$_2$ have indicated[2,12] that below T_N the magnetic ground state is separated from the spin excitations by a gap of 2 meV at the magnetic zone center. This gap is associated with a longitudinal excitation between the two lowest-lying f levels, which in this case are singlets. In such a system the magnetic moments must be induced self-consistently by the mixing of the f levels by the exchange field. This induced-moment picture can account, at least qualitatively, for the very small value of the moment at saturation. It also might account for the unusual temperature dependence of the order parameter provided that there are ungapped regions on the Fermi surface below T_N. The ungapped regions would provide the finite density of thermally accessible states that would limit the growth of the elastic magnetic component relative to that in a localized moment system (e.g., UAs). Specific-heat measurements have shown that at least one-third of the Fermi surface is ungapped.[5] We point out that this behavior is quite different from what is observed in a clean BCS superconductor or an Ising magnet, for example, where there is an isotropic gap and the order parameter saturates by approximately $T_N/2$.

It is interesting to note the similarities between the URu$_2$Si$_2$ results described above for both x rays and neutrons and the neutron studies in UPt$_3$.[4] First, in UPt$_3$ the magnetic Bragg intensity grows linearly over an extended range from $T_N = 5$ K to $T_N/10$. In addition, the very small saturation moment of $0.02\mu_B$ in UPt_3 below $T = 0.5$ K is complemented by a large quasielastic fluctuating density of states. However, for URu_2Si_2 the strong spin fluctuations ($\mu_{sf} \cong 2\mu_B$) are mostly sharp spin excitations. This lends support to the idea described above that the slow linear rise of the order parameter is controlled by the ungapped portion of the Fermi surface and not by the damping of the spin excitations.

The sharp onset of long-range antiferromagnetic order that we have observed at $T_N = 17$ K corresponds to the large λ anomaly in the specific heat and the development of propagating, longitudinal spin waves observed with inelastic neutron scattering.[2] This fact, in conjunction with the relatively long-range, temperature-independent correlation length of Fig. 3 (probably limited in size by crystal defects), characterizes the static magnetic order in URu_2Si_2 as intrinsic. Furthermore, the persistence of the long-range antiferromagnetic order into the superconducting phase strengthens the claim that these two ground states coexist on a microscopic scale.

In conclusion, we have applied x-ray-resonance magnetic scattering to study the antiferromagnetic order of the very small moment in URu_2Si_2. These studies have revealed the correspondence between the large λ anomaly in the specific heat and the antiferromagnetic transition, as well as long correlation lengths, in contrast to earlier neutron measurements. Recent neutron measurements of the ordered moment in a sample cut from the same boule that we have measured agree with our results, indicating that sample quality plays an important role in the static magnetic order of this system. The unusually broad range of growth of the ordered moment below T_N indicates the influence of an ungapped region. Finally, our results clearly demonstrate the coexistence of antiferromagnetism and superconductivity below $T_c = 1.3$ K, and that no change in the ordered moment is observed below T_c, to within the systematic errors of this experiment.

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