Neutron-Scattering Measurements of Long-Range Antiferromagnetic Order in URu_2Si_2

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Neutron scattering from a high-quality single crystal of the heavy-fermion superconductor URu_2Si_2 has shown the onset of the small, antiferromagnetic ordered moment ($0.037\mu_B$) occurs abruptly at T_N, unlike the gradual onset in previously studied samples. The results are in good agreement with x-ray-resonance magnetic scattering. The magnitude of the moment and the magnetic excitation spectrum are the same as previously observed, including the anomalous damping of excitations with a component of momentum transfer along the c^* axis. This shows that these are intrinsic properties of URu_2Si_2 and confirms the microscopic coexistence of antiferromagnetism and superconductivity.

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The persistence of antiferromagnetic order below the superconducting transition temperature ($T_c \simeq 1.2$ K) in the heavy-fermion compound URu_2Si_2 (Refs. 1-4) has led to a great deal of speculation about the nature of the superconducting state. Although the experimental evidence does suggest coexistence of superconductivity with antiferromagnetism there are aspects of the antiferromagnetic ordering that occurs below $T_N \simeq 17$ K which remain puzzling. Neutron-scattering measurements have shown that the ordered moment in this system is extremely small, about $0.04\mu_B$.[2,3] In addition, the magnetic Bragg peaks associated with the antiferromagnetic order are broad along the c axis of the body-centered-tetragonal lattice[3] indicating that there is only short-range order in this direction. In the superconducting phase, the magnetic-Bragg-peak intensity exhibits unusual nonequilibrium behavior in the presence of a magnetic field that may be due to domain effects.[4]

The temperature dependence of the antiferromagnetic order parameter is also anomalous. The Bragg-peak intensity decreases substantially as T_N is approached from below but does not go to zero at 17 K.[3] There is significant intensity even above 20 K which cannot be attributed to critical scattering. This is in contrast to the bulk properties of URu_2Si_2 which, in smaller samples, show distinct anomalies at T_N in the specific heat and resistivity.[1] Unlike the elastic component of the magnetic neutron scattering, the excitation spectrum does change dramatically at T_N, from an overdamped response to a sharp crystal-field-like response with an overdamped higher-energy component.[2,3]

Another heavy-fermion superconductor that orders antiferromagnetically is UPt_3.[5] This material also has an extremely small ordered moment ($0.02\mu_B$) and Bragg peaks that are not resolution limited. In both cases the failure to observe infinitely long-range antiferromagnetic order raises the question of whether there is microscopic coexistence of antiferromagnetism and superconductivity, or, if only small regions of the crystal are antiferromagnetic, perhaps associated with crystalline imperfections or impurities. The question of coexistence is particularly important since antiferromagnetic spin fluctuations are thought to play an important role in mediating the superconducting pairing in the heavy-fermion superconductors.[6] This is especially true in the case of UPt_3 where the complicated superconducting phase diagram has been explained by a coupling between the superconducting and antiferromagnetic order parameters.[7]

We have carried out neutron-scattering measurements on a new, high-quality single crystal of URu_2Si_2 grown from the higher-purity uranium than previous samples. The measurements were carried out on the L3 triple-axis spectrometer at the NRU reactor, Chalk River. Unlike the previous measurements, the magnetic Bragg peaks are resolution limited, indicating long-range antiferromagnetic order. In addition, the measurements were carried out on a crystal cut from the same boule as the sample used for x-ray-resonance magnetic scattering (XRMS) measurements of Isaacs *et al.*[8] The good agreement between these two probes confirms the usefulness of XRMS as a tool for studying these extremely small-moment systems.

The crystal structure of URu_2Si_2 is the body-centered-tetragonal $ThCr_2Si_2$ structure with $a = 4.124$ Å and $c = 9.582$ Å at 4.2 K. A 0.3-cm^3 crystal was cut from a cylindrical boule about 2 cm long by cleaving with a razor blade perpendicular to the c axis. The single crystal from which the sample was cut was grown by a modified Czochralski method that has been described elsewhere.[9] Ultrahigh-purity uranium from Ames Laboratory was used for the growth. Samples with this special iron-free uranium have been found to have somewhat higher superconducting transition temperatures[9] than those without[1] ($T_c = 1.33$ K rather than 1.2 K).

The crystal was mounted in a pumped ^{4}He cryostat in the (hhl) scattering plane. The temperature was monitored using a Si resistor. For the elastic-scattering measurements the (002) reflection from pyrolytic graphite (PG) was used to monochromate and analyze the neutron beam. The neutron energy was 1.2 Thz. A cooled Be filter was placed in the incident neutron beam to remove higher-order contamination. The inelastic-scattering measurements were carried out using a Si(111) monochromator and a PG (002) analyzer. A fixed final neutron energy of 3.52 THz was used with a PG filter in the scattered beam to reduce higher-order contamination. Collimation before and after the sample was 0.6° and 0.8°, respectively, in both configurations.

Figure 1 shows a series of scans through the (111) antiferromagnetic Bragg peak at $T = 2.7$, 11, and 15 K. The contribution from higher-order nuclear scattering as well as the flat instrumental background was determined by scans at 19 and 30 K (above T_N) and were subtracted from the data. The (111) reflection has less higher-order contamination than the (100) reflection used in previous work because the (222) and (444) reflections [which give rise to $\lambda/2$ and $\lambda/4$ scattering at the (111) position] are much weaker than (200) and (400). The sum of the flat background and higher-order peaks represented about 10% of the intensity at 2.7 K. The lines are the results of least-squares fits with Gaussian functions. The data of Fig. 1 along with similar scans at other temperatures are resolution limited along the (110) direction. The same is true along the (001) direction.

Figure 2 shows the temperature dependence of the integrated intensity determined from the least-squares fits to data of the type shown in Fig. 1. The solid circles are the neutron-scattering results and the open circles are the x-ray-scattering results of Isaacs *et al.*[8] (scaled to agree at 4.2 K). The two probes are in good agreement indicating that the x-ray-scattering intensity scales with the square of the order parameter over the range of intensity covered by the measurements. The antiferromagnetic-Bragg-peak intensity vanishes approximately linearly as T_N is approached from below suggesting mean-field behavior ($\beta = \frac{1}{2}$), possibly due to the long-range nature of the RKKY interactions responsible for ordering in the material.[3] There is a slight rounding of the transition near 17 K reminiscent of that seen in the earlier work[2,3] but much less pronounced. There is no temperature-dependent contribution above 19 K.

The size of the ordered moment can be determined by normalizing the antiferromagnetic-Bragg-peak intensity to a nuclear peak. Comparison with the (110) and (002) nuclear reflections yields a value of $(0.037 \pm 0.005)\mu_B$ per uranium atom. Both of the nuclear peaks are relatively weak so extinction should not be a major factor. The (110) reflection is quite close to (111) so resolution effects should be the same for these two peaks. The fact that normalizing to (110) and (002) gives the same ordered moment suggests that resolution and multiple-scattering corrections are not significant. The value ob-

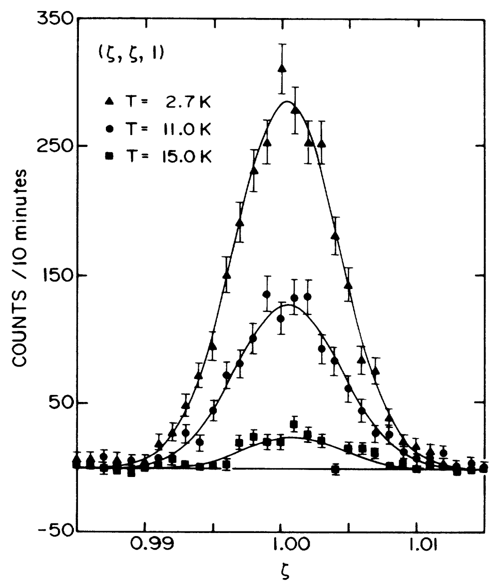

FIG. 1. Scans through the (111) antiferromagnetic Bragg peak at $T = 2.7$, 11, and 15 K. The background determined at temperatures above T_N has been subtracted. The lines are the results of least-squares fits by Gaussian functions.

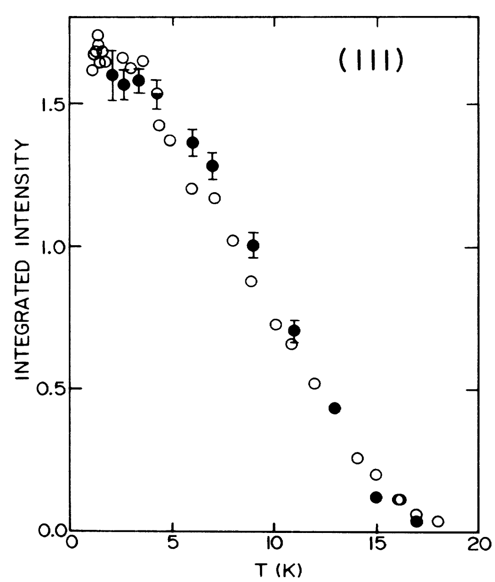

FIG. 2. The temperature dependence of the integrated intensity of the (111) magnetic Bragg peak. The solid circles are the neutron-scattering results and the open circles are the x-ray-scattering results from Ref. 8.

tained for the ordered moment is the same as previously reported,[2,3] indicating that although the correlation length is significantly longer in this new sample the size of the ordered moment remains extremely small.

One possible explanation for the existence of long-range order in the new crystal is that the use of high-purity uranium reduces the incidence of stacking faults common in this class of materials.[10] The possible importance of these stacking faults is suggested by the fact that in the earlier work the short correlation length was observed principally along the c axis.[3] Another possibility is that silicon stoichiometry has an effect on the antiferromagnetic order. Experience gained in growing other UT_2Si_2 materials (with T = Ni and Ir) has shown that the silicon content of the crystal decreases along the growth direction due to the evaporation of silicon from the melt. There is consequently a decrease in the unit-cell volume along the growth direction of typically 0.5 Å^3. Guinier photographs of material from either end of the boule from which the present sample was cut gave unit-cell volumes of 163.4(1) and 163.5(1) Å^3, indicating a more uniform silicon concentration. This is difficult to control but the present results suggest that a detailed study of the importance of this effect might prove fruitful.

Whatever sample-dependent effect is responsible for the short-range order and anomalous temperature dependence of the order parameter observed in earlier work has not been entirely eliminated from the crystal used in the present study. The effect is still seen in the slight rounding of the intensity around 17 K in Fig. 2. There is also a small intrinsic width in the magnetic Bragg peak not observable in these measurements but visible in the higher-resolution x-ray-scattering measurements.[8] It now seems likely that these effects are not features intrinsic to URu_2Si_2. This observation suggests that the finite correlation length observed in UPt_3 (Ref. 5) is also due to crystalline defects.

The magnetic excitation spectrum is not sensitive to the crystal quality. Inelastic-neutron-scattering measurements give the same results as previous work;[2,3] the magnetic excitations within the basal plane are sharp and dispersive but there is a substantial damping that sets in with increasing component of momentum transfer along the c axis. This is shown in the constant-Q scans of Fig. 3 at (1,1,1.10), (1,1,1.15), and (1,1,1.30). The sharp mode seen in the scan at (1,1,1.10) acquires a high-energy tail as the c component of Q is increased to 1.15 and is completely overdamped for Q_c equal to 1.30. This qualitative behavior is the same as previously observed as are the excitation energies throughout the zone. A possible explanation of the anomalous damping of the magnetic excitations with components of wave vector along c was that it resulted from the interaction of these excitations with stacking faults. These new results are important as they show the same anomalous damping in

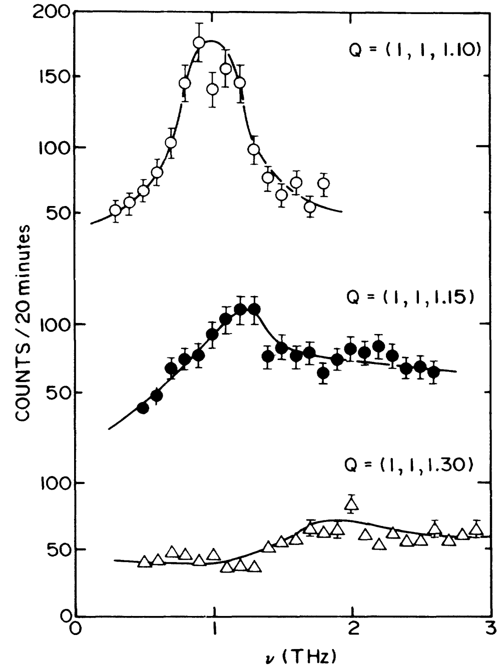

FIG. 3. Constant-Q scans at T = 4.2 K for Q = (1,1,1.10), (1,1,1.15), and (1,1,1.30). There is a substantial increase in the damping with increasing momentum transfer along the c axis. The lines are guides to the eye.

a higher-quality crystal, strongly supporting the picture of this dynamic effect as intrinsic to the pure heavy-fermion material.

In conclusion, the present work shows that, in contrast to earlier measurements,[2,3] the antiferromagnetic order in URu_2Si_2 is long range with a sharp onset. The reduced correlation length seen previously is attributed to sample-dependent effects that do not change the magnitude of the ordered moment or the magnetic excitation spectrum. Although the correlation length in the new sample is much longer, the size of the ordered moment remains the same, $(0.037 \pm 0.005)\mu_B$; it is, therefore, an intrinsic feature. The results obtained are in good agreement with XRMS measurements[8] on a sample cut from the same boule as the neutron sample confirming that XRMS is capable of probing extremely small moment systems. The existence of long-range antiferromagnetic order in this new sample of URu_2Si_2 clearly shows that there is microscopic coexistence of antiferromagnetism and superconductivity in this heavy-fermion material.

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