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Neutron Scattering Studies on Heavy Fermion Superconductors

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We review recent neutron scattering studies on uranium-based heavy fermion superconductors. The coupling between magnetic and superconducting order parameters was observed in UPt_3, UPd_2Al_3, URu_2Si_2, and UNi_2Al_3. In UPd_2Al_3, we observed a low energy response strongly related to the superconductivity. These results are indicative of the strong interplay between magnetism and superconductivity. We also report the unusual behaviors of the weak antiferromagnetic ordering, the long range magnetic correlation in UPt_3 at ultra-low temperatures, and the pressure induced magnetic transition from the weak (0.02 μ_B/U) to a high moment state (0.4 μ_B/U) at 1.5 GPa in URu_2Si_2.

KEYWORDS: neutron scattering, heavy fermion superconductors

§1. Introduction

The unconventional superconductivity and the interplay between magnetism and superconductivity in heavy fermion superconductors attracts much interest.[1] Recently the neutron scattering group in JAERI concentrates on the study of heavy fermion superconductors with collabolation of many groups, especially Onuki Lab. in Osaka Univ. and JAERI-ASRC. In this paper we review our recent neutron scattering studies on heavy fermion superconductors.

§2. Experimental

Most of the neutron scattering experiments were carried out using a cold and thermal neutron triple-axis spectrometers LTAS and TAS1, respectively, installed at the research reactor JRR-3M in Japan Atomic Energy Research Institute (JAERI). Special attention has been paied for the shielding of the fast neutron from samples to reduce the background. For high pressure experiments a tiny sample (4ϕ x10 mm) was encapsulated in a McWhan type cell and compressed with hydrostatic pressure up to 3 GPa.

§3. Result and Discussion

Figure 1 is the temperature dependence of the AFM Bragg intensity in UPd_2Al_3,[3,4] URu_2Si_2,[5] UNi_2Al_3,[6] and UPt_3.[7,8] In these compounds we observed a clear decrease of the AFM Bragg intensity below the superconducting transition temperature T_C. This is evidence for the coupling of the magnetic and superconducting order parameters. This phenomenum is first discovered in UPt_3.[9] The magnetic ordering in UPt_3 and URu_2Si_2 is quite unusual. The moment size is extremely small and the magnetic correlation remains finite. It is considered to be the dynamical fluctuation, not the static ordering. On the other hand, in UPd_2Al_3 the AFM ordering is rather usual with a relatively large moment (0.85 μ_B/U). We observed resolution-limited magnetic peaks below the Néel temperature of 14.5 K. From our

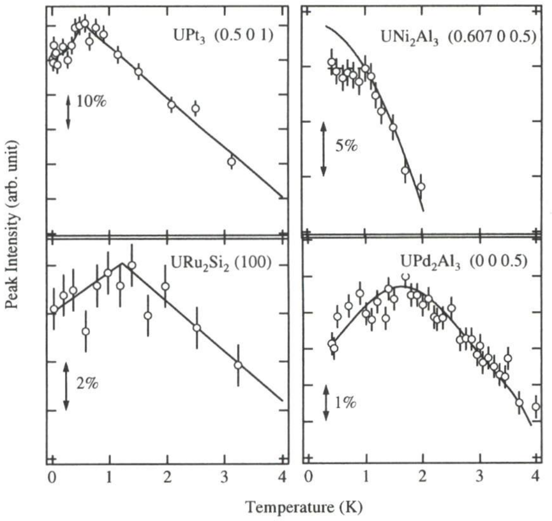

Fig. 1. Temperature dependence of the AFM Bragg Intensity for uranium heavy fermion superconductors.

results it is revealed that the coupling of the magnetic and superconducting order parameter would be a common feature in heavy fermion superconductors.

Figure 2 is the low energy spin excitation spectra of UPd_2Al_3 at an AFM Bragg point (0 0 0.5). In the normal state ($T > T_c$) we observed a quasielastic and inelastic response with the excitation energy ΔE of about 1.5 meV. The inelastic component corresponds to the spin wave excitation reported before. The quasielastic component becomes weaker with decreasing temperature. Below T_c, however, the intensity increases and the peak position shifts towards a higher energy. At $T = 0.4$ K the profile can only be explained by inelastic lineshape. We found the spin excitation gap $\Delta E = 0.36$ meV by least square fitting with Lorentzian convoluted with the resolution function and Bose factor. The obtained line width was about 0.2 meV. Furthermore, we found that the gap disappears with applying magnetic field higher than the critical field H_{c2}. The superconducting gap appearing in spin excitation was found for the first time in this com-

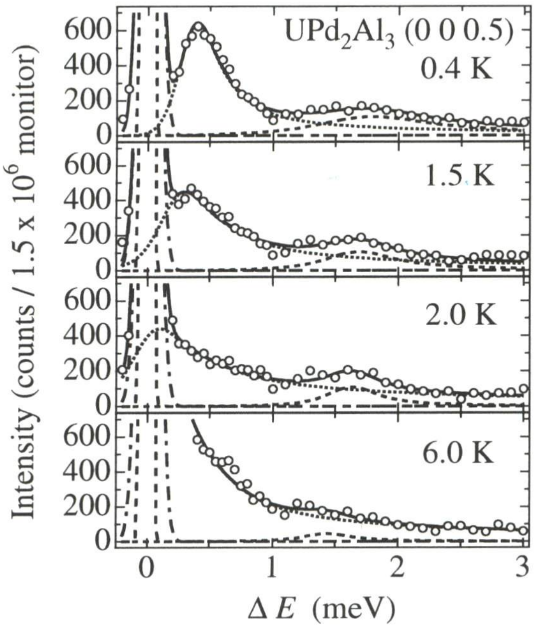

Fig. 2. Temperature dependence of the spin excitation spectra at (0 0 0.5) AFM zone center in UPd_2Al_3.

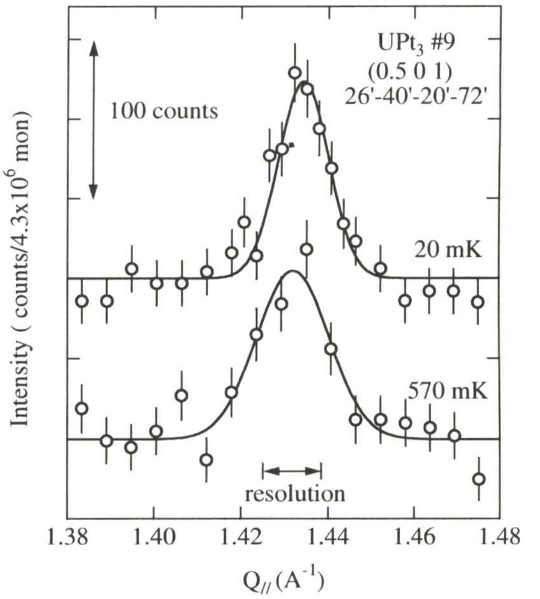

Fig. 3. The longitudinal scattering profile of the (0.5 0 1) magnetic peak in UPt_3.

pound and it is the most important feature for the spin dynamics in UPd_2Al_3. This low energy response was discovered by Sato[10] and Bernhoeft *et al.*[11] According to the analysis of their neutron and the other data relevant sheet(s) of f-electron fermi surface has been proposed.[12]

The unusual nature of the weak AFM ordering in UPt_3 and URu_2Si_2 has been one of the most mysterious problems in heavy fermion superconductors. In these systems the magnetic moment is very small, in the order of the 0.02 - 0.04 μ_B/U. The linewidth of the magnetic peak is not resolution limited, implying a finite spin correlation length. In UPt_3 no evidence of the magnetic ordering has been reported by susceptibility,[13] NMR[14] and specific heat[15] measurements, despite clear signature of the ordering in neutron[9] and magnetic X-ray[16] scattering. Therefore this magnetic correlation is suspected to be a dynamical fluctuation which has a characteristic time scale between NMR and neutron. On the other hand, it has been reported that the specific heat,[17] magnetization,[18] and thermal expansion[19] exhibit a clear anomaly at 20 mK. It would be a signature for a static ordering.

In order to clarify the nature of weak AFM of UPt_3, we have carried out ultra-low temperature neutron scattering experiments.[7,8,20] Figure 3 is the temperature dependence of the (0.5 0 1) AFM Bragg peak. The AFM peak is observed even in the high quality sample with RRR (residual resistivity ratio) = 700. Furthermore the estimated magnetic moment of 0.035 μ_B/U showed no systematic dependence on sample quality. Therefore this magnetic ordering is concluded to be an intrinsic property of UPt_3. For all samples a remarkable narrowing of the linewidth has been observed below 50 mK. A resolution limited peak was observed at 20 mK. Above 50 mK the correlation length was typically 300 Å to 700 Å, depending on the sample quality; the spin correlation length becomes longer with increasing the mean free path. Below 50 mK the correlation length increases very rapidly, and diverges at 20 mK. This temperature is in good agreement with the one of the specific heat anomaly.[17] Therefore, we can conclude that the specific heat anomaly is related to the AFM ordering in UPt_3. The narrowing of the linewidth would be a precursive dynamical slowing down towards static ordering.

In URu_2Si_2 a cusp like behavior in the resistivity is reminiscent of the CDW/SDW transition.[21] A large jump is observed in specific heat data,[22] which is extremely larger than the entropy expected from moment size. Therefore hidden order parameter like quadrapole[23,24] has been proposed as a primary order paramener. High field neutron scattering experiments reported the suppress of the magnetic order under high field, remaining order with a primary order parameter. Up to now no evidence of the orbital ordering has been obtained from resonance x-ray scattering studies. The origin of the weak AFM ordering remains an open question. The problem concerning hidden order in URu_2Si_2 is summarized in a paper.[25]

Figure 4 shows the transverse scan of the (100) magnetic Bragg peak at 1.5 K for various pressure. We found a dramatic increase of the peak intensity with pressure. From these data we found that the moment size increases continuously from 0.02 μ_B/U at $p = 0$ to 0.2 μ_B/U at 1.3 GPa, and exhibits a small jump to 0.4 μ_B/U at $p_c = 1.5$ GPa, indicative of the first order transition. For $p > 1.5$ GPa the moment decreases gradually with pressure.[26]

We have carried out the inelastic scattering experiments at high pressures.[2,27] The characteristic feature of URu_2Si_2 is the strong inelastic response with a gap at the AFM Bragg point below T_N.[28–30] We found that the gap of about 2 meV at $p = 0$ (1.5 K) decreases with pressure, and the inelastic response vanishes above p_c =1.5 GPa. It means that the large moment state is stabilized under high pressure at the expense of the strong inelastic response accompanying the quenching of the magnetic

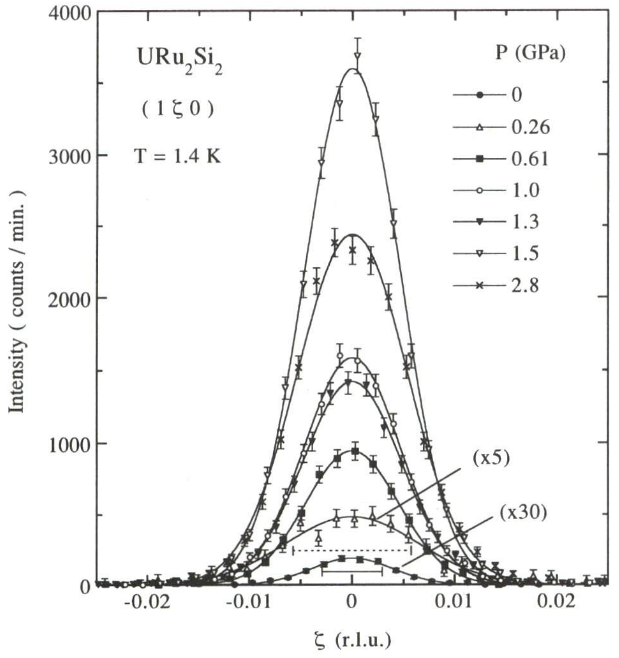

Fig.4. (100) transverse scan at 1.5 K for various pressures in URu_2Si_2.

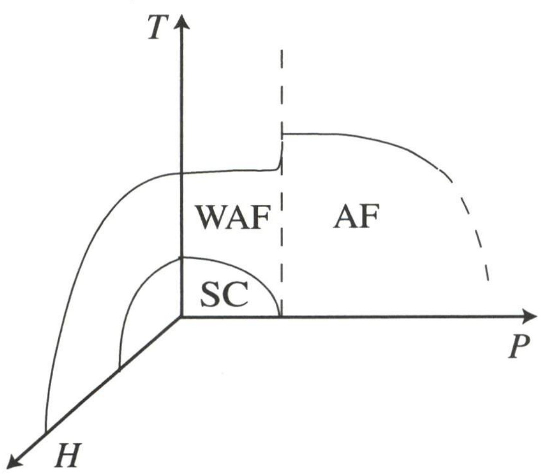

Fig.5. The phase diagram of URu_2Si_2.

excitation gap.

The high sensitivity of magnetic moment to pressure rules out the possibility that the weak AFM ordering would have a nesting origin. No large modification of the Fermi surface is expected with such a small change in lattice constant. This ordering has been reported to be static within the resolution of neutron spin echo spectroscopy,[31] while the observation of no change in Knight shift in NMR[32] is quite strange. We found that the temperature dependence of the magnetic moment looks quite different; 3D ising like behavior is observed above p_c, while a gradual increase of the peak intensity would be a nature of dynamically fluctuating AFM correlation.

Figure 5 is the phase diagram in URu_2Si_2. It should be noted that the superconducting transition temperature decreases with pressure, and the superconductivity vanishes above 1.6 GPa,[33] which is very close to p_c mentioned above. Therefore the weak AFM ordering would be essential for the coexisting heavy fermion superconductivity in URu_2Si_2. Our results remind us a crossover from the weak to large AFM state in $U(Pt_{1-x}Pd_x)_3$.[34] With substituting Pt by small amount of Pd, a strong magnetic ordering takes place, and hence the existence of dynamically fluctuated state is deduced in a pure system.[35] The phase diagram in URu_2Si_2 is very similar to the one of $U(Pd_xPt_{1-x})_3$. In both system a large moment state exists near weak AFM state which coexists with superconductivity.

§4. Conclusion

Systematic neutron scattering experiments have been carried out on uranium-based heavy fermion superconductors. Clear evidence for the strong interplay between magnetism and superconductivity has been reported, which is the most important issue for this system.

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