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Neutron Scattering Studies of the Heavy Fermion Superconductors

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ABSTRACT

Recent neutron scattering measurements of the heavy Fermion superconductors are described. These materials offer an exciting opportunity for neutron scattering since the f-electrons, which couple directly to magnetic scattering measurements, seem to be the same electrons which form the superconducting state below T_c. In addition, studies of the magnetic fluctuations in these, and other heavy Fermion systems, by inelastic magnetic neutron scattering can provide information about the nature of the low temperature Fermi liquid character of these novel compounds.

I. INTRODUCTION

The discovery of superconductivity in heavy fermion systems has generated a great deal of excitement and speculation concerning the normal and superconducting properties of these materials [1-8]. These systems are characterized by large electronic specific heat coefficients (γ) at low temperature which imply an effective mass for the f-electrons which is typically 2-3 orders of magnitude greater than the free electron mass. Large entropy changes at the superconducting transition indicate that these same f-electrons are responsible for the superconductivity. Near room temperature the heavy fermion systems exhibit Curie-Weiss behavior with large ($> 2\mu_B$) effective moments. However, at low temperature the susceptibilities deviate from this simple dependence and remain large but finite. One of the interesting questions, still unanswered, concerns the mechanism which alters the Curie-Weiss behavior of the susceptibility.

II. NEUTRON SCATTERING MEASUREMENTS

What information do we gain from neutron scattering measurements? Consider the magnetic scattering cross-section from an isotropic paramagnet [9]

$$\frac{d^2\sigma}{d\Omega dE} = \gamma_0^2 \frac{k_f}{k_i} f^2(Q) S(Q,E) e^{-2W(Q)} \qquad (1)$$

Here $\gamma_0{}^2$ is the neutron-electron dipole coupling constant, k_i and k_f are the incident and final neutron wavenumbers respectively, and $e^{-2W(Q)}$ is the Debye-Waller factor.

The physics of interest is contained in the magnetic form factor f(Q), which can be related to the wavefunction of the electrons responsible for the magnetic scattering, and S(Q,E) which is the dynamical structure factor related to the response function of the system. S(Q,E) may be in turn written as:

$$S(Q,E) = \langle n(E) + 1 \rangle \, \mathrm{Im}\chi(Q,E) \qquad (2)$$

where $\langle n(E) + 1 \rangle$ is the Bose occupation factor, $[1 - e^{-E/kT}]^{-1}$, and $\mathrm{Im}\chi(Q,E)$ is the imaginary part of the susceptibility. If the response of the system is purely relaxational and all of the Q-dependence is well described by the magnetic form factor, then

$$\mathrm{Im}\chi(Q,E) \rightarrow \mathrm{Im}\chi(E) = \frac{A\Gamma E}{\Gamma^2 + E^2} \qquad (3)$$

where Γ is the half-width at half-maximum of the quasielastic response. From equations (2) and (3) it is easily seen that at low temperature ($kT \ll \Gamma$) the scattered intensity peaks at $E = \Gamma$, while at high temperature ($kT \gg \Gamma$) the scattering becomes more symmetric about $E = 0$. The real part of the susceptibility may be obtained from the imaginary part through a Kramers-Kronig relation. This then allows a direct comparison between the local susceptibility as measured by neutron scattering, and the measured bulk susceptibility.

III. ELASTIC SCATTERING MEASUREMENTS: SUPERCONDUCTIVITY

The heavy fermion superconductors present an exciting opportunity for neutron scattering studies. Almost all of the past neutron scattering investigations have probed the superconductivity either through the electron-phonon interaction [10], or the interaction between the magnetic moment carried by one constituent and the superconducting electrons derived from another [11, 12]. In the case of the heavy fermion systems, the same f-electrons responsible for the large susceptibility are believed to also form the superconducting state below T_c. Therefore elastic and inelastic magnetic scattering measurements can directly probe the superconducting state in these systems.

The magnetic form factor, f(Q), may be obtained using polarized neutron scattering techniques, from the moment induced in a material when a magnetic field is applied. One measures the ratio of the Bragg peak intensities for neutron spin orientations parallel and antiparallel to the applied magnetic field. This polarization ratio is given by,

$$R = \frac{(F_N + F_m(Q))^2}{(F_N - F_m(Q))^2} \simeq 1 + 4\,\frac{F_m(Q)}{F_N}; \; \vec{Q} = \vec{\tau} \qquad (4)$$

F_N is the nuclear structure factor, and $F_M(Q)$ is the induced magnetic moment structure factor which is proportional to $\chi(Q,0)$, the static Q-dependent susceptibility. Figure 1 shows the induced moment structure factor of $CeCu_2Si_2$ as measured by Stassis et al. [13] at 4.2K, which is consistent with the theoretical 4f form factor, f(Q), of a Ce^{3+} ion. The static susceptibility, $\chi(0,0)$, is obtained by extrapolating the measurements to Q=0, and is in good agreement with the bulk susceptibility value. Therefore it can be concluded that the induced magnetization is of predominantly 4f character. The 5f-electron character of the magnetic scattering from UBe_{13} [14, 15] and UPt_3 [14, 16] has likewise been established. No indication of s- or d-electron contributions was observed for the momentum transfer range measured.

According to the BCS theory, the spin contribution to the susceptibility should vanish exponentially below T_c, as T approaches 0^oK, due to spin-pairing of the superconducting electrons. The temperature dependence of the paramagnetic susceptibility in a superconductor was calculated by Yosida [17], and is in good agreement with neutron diffraction studies of the induced susceptibility in V_3Si by Shull and Wedgwood [18]. It has been proposed that the superconducting ground state of the heavy Fermion systems is not the conventional s-wave BCS pairing. For triplet (p-wave) pairing one might expect qualitatively different behavior in the temperature dependence of the induced moment as measured by neutron scattering. Extending the analogy between the heavy Fermion systems and superfluid ^3He [19], a superconducting state corresponding to the A-phase (ABM-state) would exhibit no change in the electronic spin susceptibility below T_c, while a B-phase (BW-state) description would exhibit a reduced suppression of the susceptibility.

Very recent measurements [14] of the induced moment magnetic form factor of UBe_{13} found no difference in the induced moment below T_c. An additional temperature dependent contribution for UPt_3 was tentatively identified as arising from the polarization of the Pt nuclei. While one may be tempted to cite the results of the induced moment measurement of UBe_{13} as evidence of an exotic pairing mechanism for the superconducting electrons in this material, the relative importance of spin-orbit coupling in this system must first be addressed.

IV. INELASTIC NEUTRON SCATTERING: MAGNETIC FLUCTUATIONS

Inelastic neutron scattering measurements can be used to probe the energy dependence of magnetic fluctuations in a system, providing information about the characteristic energy scale of the fluctuations, crystal field effects, and collective excitations through the Q-dependence of the inelastic spectra.

Horn et al. [20] have measured the neutron energy loss spectrum of $CeCu_2Si_2$ and observed a quasielastic response with a strongly temperature dependent half-width (Γ) which at 10K was about 1 meV. Other inelastic peaks observed in the spectra at 12.5 meV and 31 meV were attributed to crystal field excitations. The measured local susceptibility was found to be significantly larger than the bulk susceptibility, and this difference was attributed to correlations between the 4f-electrons of Ce^{3+} and the conduction electrons mediated by "Kondo-type" spin fluctuations. However recent measurements [13, 21] found good agreement between the measured local susceptibility and bulk values, while the measured half-width of the quasielastic scattering at low temperature was in basic agreement with the previous study.

The magnetic inelastic spectrum of UPt_3 measured by Aeppli et al. [16] using polarized neutrons is shown in the top panel of Fig. 2. The data correspond to the number of spin-flip events observed with the neutron polarization oriented parallel (HF) and perpendicular

(VF) to the momentum transfer, and yield one-half of the magnetic cross-section (Eqn.(1)) free of contamination from nuclear contributions. The magnetic spectrum of UPt_3 is characterized by a quasielastic response with $\Gamma \approx 10meV$. The local susceptibility determined from this investigation is in good agreement with the low temperature bulk value.

Polarized and unpolarized neutron scattering studies of UBe_{13} have been performed by Goldman et al. [15]. In the unpolarized neutron beam investigation the phonon contribution to the inelastic spectrum was estimated from a measurement of the non-magnetic isostructural compound $ThBe_{13}$ obtained under the same conditions, and corrected for minor differences in the scattering lengths of Th and U, and sample volume. The magnetic scattering contribution to the UBe_{13} inelastic spectrum is shown in the bottom panel of Fig. 2, and is consistent with the polarized neutron beam measurements. As was the case for UPt_3, a broad quasielastic response ($\Gamma \approx 14$ meV) at 10K is observed and, within experimental uncertainty, the measured local susceptibility nearly exhausts the low temperature bulk value.

The half-width, Γ, measured in these experiments may be interpreted as the characteristic spin fluctuation temperature for the f-electrons in these systems. The large value of γ, for the heavy Fermion systems, suggests a narrow peak in the density of states for the f-electrons at the Fermi energy [6]. It is clear however from measurements of the linewidths of the quasielastic scattering from $CeCu_2Si_2$ and UBe_{13}, which have nearly the same value of γ, that there is no simple scaling relation between γ and Γ. It is worth noting that two of the known cerium-based heavy Fermion systems, $CeCu_2Si_2$ and

$CeAl_3$ [22] (non-superconducting), both exhibit strongly temperature dependent quasielastic linewidths, which have small residual ($T \rightarrow 0$) values of $\approx$ 2 meV and $\approx$ 0.5 meV respectively. This behavior motivated the original characterization of these materials as "Kondo-like" systems. Studies of the magnetic fluctuations in another cerium-based heavy Fermion system, $CeCu_6$, are in progress. Although temperature dependence investigations of the uranium-based heavy Fermion systems have not been completed, the low temperature inelastic spectra (Fig. 2) are similar to the broad featureless response observed in several actinide systems [23]. Studies of U_2Zn_{17}, a heavy Fermion system which orders magnetically at low temperature are underway.

III. FUTURE WORK

While it is clear that neutron scattering studies of the heavy Fermion systems offer tremendous potential for unraveling the physics of these fascinating compounds, much more work on both the theoretical and experimental fronts needs to be done. At this time, a variety of heavy Fermion systems are under investigation, and it is hoped that new results will be forthcoming in the near future. First, it is of some importance to determine whether there are observable trends in the quasielastic linewidths from system to system and if, in fact, some systematics concerning Γ, (γ and χ) can be determined. Second, further studies of the Q-dependence of the inelastic magnetic scattering are planned which may yield some insight into electron-electron correlations characteristic of the low temperature Fermi-liquid-like behavior of these systems. Further studies of the heavy Fermion systems below T_c may be instrumental in understanding the character of the superconducting state of this novel class of materials.

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Figure Captions

Fig. 1. Measured induced moment structure factor $4F_M(Q)$ plotted as a function of $\sin\theta/\lambda$. The solid line was obtained by fitting the data to the 4f form factor of Ce^{3+} (after Ref 13).

Fig. 2 top: Constant $Q = 2Å^{-1}$ polarized neutron scattering spectrum obtained from UPt_3 at 1.3K. The solid line is a fit to the data using a Lorentzian lineshape modified by the Bose occupation factor as described in the text (after Ref. 16).

bottom: Constant $Q = 2\ Å^{-1}$ magnetic inelastic scattering spectrum of UBe_{13} at 10K. The solid line is again a fit to the data as described above (after Ref. 15).

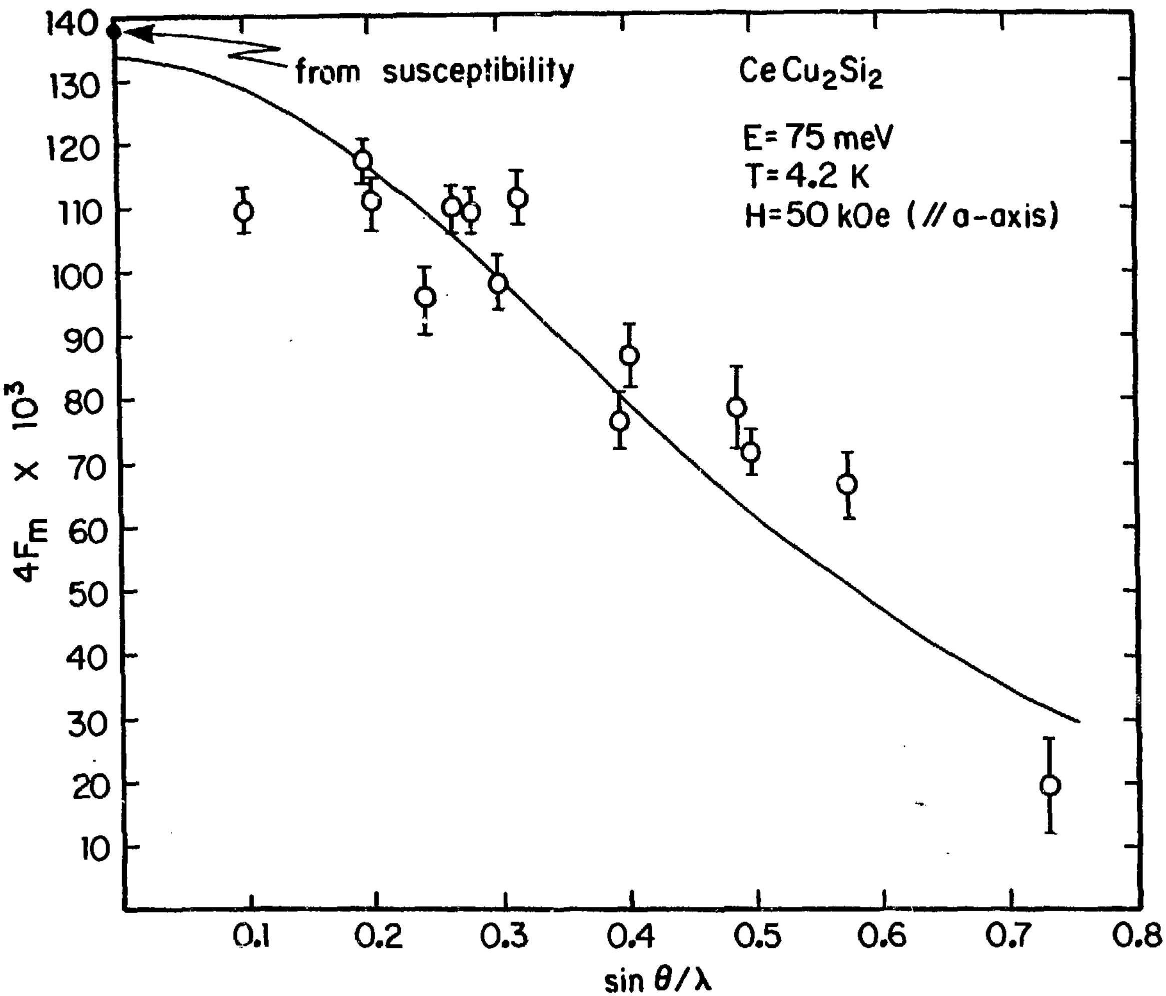
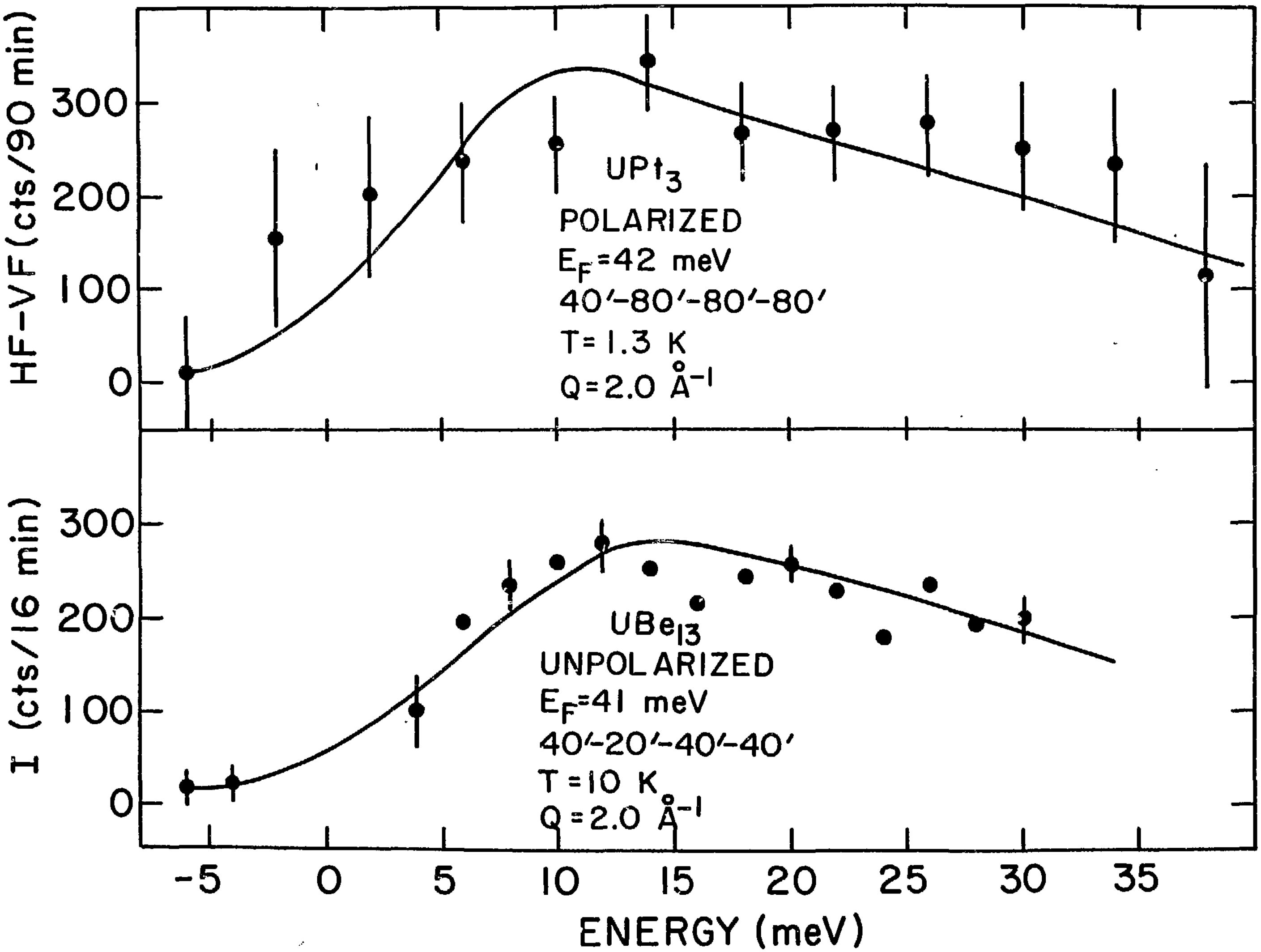

140
130
120
110
100
90
80
70
60
50
40
30
20
10
4F_m X 10^3
from susceptibility
Ce Cu2Si2
E= 75 meV
T=4.2 K
H=50 kOe (// a-axis)
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
sin θ/λ

Figure 1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300
200
100
0
UPt3
POLARIZED
EF=42 meV
40′-80′-80′-80′
T= 1.3 K
Q=2.0 Å-1
I (cts/16 min)
300
200
100
0
UBe13
UNPOLARIZED
EF=41 meV
40′-20′-40′-40′
T=10 K
Q=2.0 Å-1
-5
0
5
10
15
20
25
30
35
ENERGY (meV)

Figure 2