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Induced-moment magnetic form factor of the heavy-fermion superconductors UPt_3 , UBe_{13} , and CeCu_2Si_2

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Polarized neutron scattering techniques have been used to study the spatial distribution and temperature dependence of the magnetization induced in single crystals of UPt_3 , UBe_{13} , and CeCu_2Si_2 by an externally applied magnetic field. We find that (a) the induced magnetization in the normal low-temperature state is predominantly of f -electronic character and that (b) the generalized electronic susceptibility $\chi(\mathbf{Q},0)$ (where \mathbf{Q} is a reciprocal-lattice vector), obtained by measurements with $H < H_{c2}$, is temperature independent between 4.2 K and approximately 0.1 K. This result implies either that the superconductivity in these compounds is unconventional or that there is significant spin-orbit scattering by impurities in the samples. This latter possibility notwithstanding, the experimental results impose severe restrictions on the possible superconducting states of these systems.

I. INTRODUCTION

In recent years, the properties of heavy-electron systems have been the subject of many theoretical and experimental investigations.¹ These systems are rare-earth or actinide intermetallic compounds characterized by very large specific-heat coefficients (typically, the electronic specific-heat coefficient γ of these compounds corresponds to an effective electronic mass of the order of $100m_e$).

At high temperatures, the properties of these systems are typical of intermetallic compounds with localized f electrons; in particular, the magnetic susceptibility follows a Curie-Weiss law. At low temperatures, on the other hand, their properties are characteristic of a highly correlated electronic Fermi liquid; in particular, their magnetic susceptibility is Pauli-like.

Among these systems, those that become superconducting (i.e., CeCu_2Si_2 , UBe_{13} , and UPt_3) are of particular interest. The values of H_{c2}/T_c and the entropy change in passing through T_c have been found to be consistent with the normal-state γ , indicating that the heavy electrons are responsible for the superconductivity in these systems. This observation and the unusual superconducting-state properties of these systems have raised important questions regarding the nature of their superconducting state.

In particular, it has been suggested²⁻⁶ by analogy to ^3He , that these systems are odd-parity superconductors. Actually, a polar state has been suggested for the superconducting phase of UPt_3 .^{2,3} Both a polar⁴ and an Anderson-Brinkman-Morel (ABM) state⁵ have been proposed for UBe_{13} , and an ABM state⁶ for CeCu_2Si_2 . However, it has also been argued⁷⁻¹⁰ that these systems, in particular, CeCu_2Si_2 , may be conventional s -wave superconductors.

Information about the nature of the superconducting electrons and the superconducting state can be obtained by neutron scattering studies of the spatial distribution and temperature dependence of the magnetization induced by an externally applied magnetic field. The measurements can, in principle, determine the temperature dependence of the *spin susceptibility* below T_c , and thus provide information about the nature of the superconducting state. In fact, measurements¹¹ by Shull and Wedgwood on V_3Si showed that the d electrons are responsible for the superconductivity, and that the spin susceptibility below T_c has a temperature dependence consistent with s -wave superconductivity.¹² We were thus motivated to undertake similar studies on the heavy-electron superconductors, and in this Rapid Communication we present the results obtained from our experiments on UBe_{13} , UPt_3 , and CeCu_2Si_2 .

II. EXPERIMENT

The quantity measured in the experiments is the polarization ratio R defined as the ratio of the Bragg peak intensities for the two neutron spin orientations, parallel and antiparallel, to the applied magnetic field. If we denote by F_n and $F_m(\mathbf{Q})$ (where \mathbf{Q} is a reciprocal-lattice vector) the nuclear and induced magnetic structure factor of the crystal, respectively, then

$$R - 1 = \frac{(F_n + F_m)^2}{(F_n - F_m)^2} - 1 \approx 4 \frac{F_m(\mathbf{Q})}{F_n},$$

where in the last step it was assumed that $F_m(\mathbf{Q}) \ll F_n$, a condition fulfilled in the present experiments. Since F_n can be evaluated from the known coherent nuclear scattering amplitudes, the results obtained by measuring $R - 1$ at several reciprocal-lattice vectors determine the induced-moment form factor $F_m(\mathbf{Q})$, a quantity proportional to the static generalized susceptibility $\chi(\mathbf{Q}, 0)$. Thus, measurement of the induced-moment form factor at a given temperature provides information about the electronic wave functions at the Fermi level. On the other hand, by studying the temperature dependence below T_c of $\chi(\mathbf{Q}, 0)$ at fixed \mathbf{Q} with $H < H_{c2}$, one obtains information about the change in the spin susceptibility due to the electronic pairing in the superconducting state. For s pairing, for instance, the spin susceptibility decreases to zero at $T = 0$ following the BCS theoretical prediction.¹²

The single crystals used in the experiments were prepared at Bell Laboratories (CeCu_2Si_2), and at Los Alamos National Laboratory (UBe_{13} , UPt_3). All samples used in the experiments were found to be superconducting with T_c 's in agreement with generally accepted values. Measurements of the rocking curves of the crystals, using a Ge perfect crystal as monochromator, showed that the CeCu_2Si_2 and UBe_{13} samples were high-quality single crystals, each with a mosaic spread η between 1 and 2 min of arc; for UPt_3 , a mosaic spread of approximately 12 min of arc was measured. Although the crystals used in the experiments had cross sections of approximately $1 \times 1 \text{ mm}^2$, the secondary extinction corrections to the CeCu_2Si_2 and UBe_{13} data were not negligible. To assess this correction, the polarization ratios of some reflections [(002) and (200) for CeCu_2Si_2 , (200) and (800) for UBe_{13}] were measured at selected incident neutron energies between 14.7 and 75 meV (in these measurements a pyrolytic graphite filter was used to attenuate the second-order contamination of the incident beam). For most reflections of both crystals, the secondary extinction correction to the measured polarization ratio was of the order of 5%.

The induced-moment form-factor measurements were performed using polarized neutron spectrometers at the High Flux Beam Reactor (HFBR) of the Brookhaven National Laboratory and at the High Flux Isotope Reactor (HFIR) of the Oak Ridge National Laboratory. All measurements between 4.2 K and approximately 50 mK were performed at the HFIR using a helium dilution refrigerator. The 4.2-K data taken at the HFIR agreed, to within experimental precision, with those obtained at the HFBR. All measurements were performed at an incident neutron energy of 75 meV, since at this energy a Pu filter can be

used whenever necessary, to minimize the second-order contamination of the beam. The measurements were performed with the external magnetic field applied along the a axis for CeCu_2Si_2 and UPt_3 , and along the $[1\bar{1}0]$ direction for UBe_{13} .

The measured polarization ratios were corrected for the effect of incomplete polarization of the neutron beam by measuring the instrumental flipping ratio at each value of the externally applied magnetic field. The neutron-spin-neutron-orbit¹³ and core-diamagnetism corrections¹⁴ were estimated and found to be negligible.

In assessing the temperature dependence of the polarization ratios of various reflections, the UPt_3 and CeCu_2Si_2 data were corrected for the contribution arising from the nuclear polarization of Pt and Cu, respectively. In CeCu_2Si_2 the Cu nuclear polarization effects could be independently assessed, since in this compound there are nuclear reflections with no Cu contribution. The contribution of the Pt nuclear polarization was checked by performing measurements on samples of pure Pt. The results of these experiments were in agreement, to within experimental precision, with the calculated values.

Since no temperature dependence was observed below T_c in all three compounds, we felt that it was necessary to repeat the original Shull-Wedgwood experiment on V_3Si as an overall check of the experimental procedure. The results obtained on V_3Si were found to agree, to within experimental precision, with the results¹¹ of Shull and Wedgwood.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The induced-moment form factors of UPt_3 and UBe_{13} were obtained, at 4.2 and 4.5 K, respectively, by measuring the polarization ratios of several reflections in an applied field of 50 kOe. The results are summarized in Figs. 1 and 2. It can be seen (Figs. 1 and 2) that the data can be fitted quite well by the theoretical $5f$ form factor of the U^{3+} ion. Furthermore, the static susceptibilities obtained by extrapolating the measured form factors to $\mathbf{Q} = 0$, are

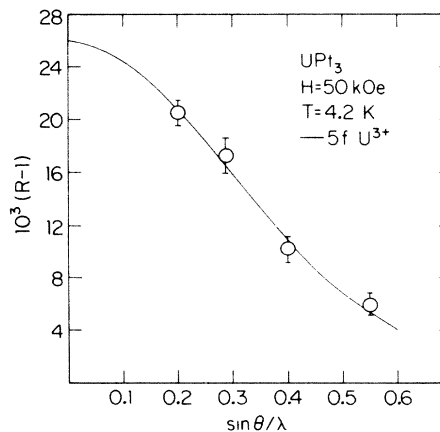


FIG. 1. Induced-moment form factor of UPt_3 in the normal low-temperature state ($T = 4.2 \text{ K}$, $H = 50 \text{ kOe}$). The solid line was obtained by fitting the data to the $5f$ form factor of U^{3+} ion.

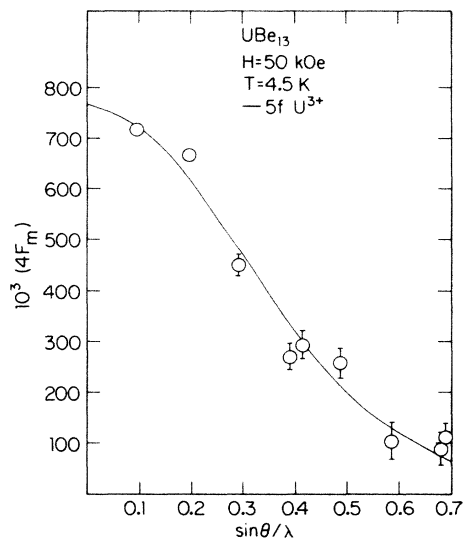


FIG. 2. Induced-moment form factor of UBe_{13} in the normal low-temperature state ($T = 4.5$ K, $H = 50$ kOe). The solid line was obtained by fitting the data to the $5f$ form factor of U^{3+} ion. The size of the circles for the first two reflections indicate the experimental precision.

in reasonable agreement with bulk susceptibility measurements. The measured form factor of CeCu_2Si_2 at 4.2 K was also found¹⁵ to be in good agreement with the theoretical $4f$ form factor of the Ce^{3+} ion. We therefore conclude that the induced magnetization in UPt_3 , UBe_{13} , and CeCu_2Si_2 at 4.2 K is predominantly of f -electron character. This result is consistent with a Fermi-liquid description (with high f -electron density at E_F) of the normal low-temperature state of these systems.

The temperature dependence of $\chi(\mathbf{Q},0)$ below 4.2 K and through the superconducting transition temperature was determined by measurements in magnetic fields lower than H_{c2} . Detailed measurements were performed in 14 and 6 kOe on UPt_3 , in 6 kOe on CeCu_2Si_2 , and in 6 kOe on UBe_{13} . The polarization ratios of several reflections were studied down to approximately 50 mK for UPt_3 , 140 mK for CeCu_2Si_2 , and 100 mK for UBe_{13} . The experimental results for some reflections are summarized in Figs. 3–5. To within experimental precision, we do not observe any change in $\chi(\mathbf{Q},0)$. For UBe_{13} , this result is consistent with NMR experiments⁴ in which no ^9Be NMR shift below T_c was observed in a field of 15 kOe. On the other hand, this result is in contradiction with the striking decrease of the Knight shift below T_c that was observed, in a field of 5 kOe, in recent μ^+ spin-rotation experiments.¹⁶

The absence of any significant temperature dependence in $\chi(\mathbf{Q},0)$ below T_c is compatible with s -wave superconductivity only if one assumes that the spin-orbit scattering rate by impurities in the samples is so large as to significantly reduce^{17,18} the superconducting correlations between electrons. This possibility notwithstanding, the experimental results imply that the superconducting state in these compounds is unconventional. Actually, the absence of any significant temperature dependence in $\chi(\mathbf{Q},0)$ below T_c imposes severe restrictions on the possible superconducting states of these systems.

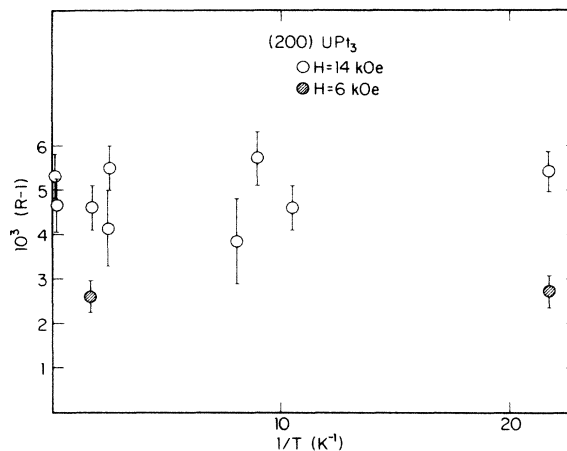


FIG. 3. Temperature dependence of the residual polarization ratio $(R - 1)$ of the (200) reflection of UPt_3 for applied fields of 12 and 6 kOe.

The superconducting classes of these systems have been obtained^{19–24} by group theoretical techniques for cubic (UBe_{13}), hexagonal (UPt_3), as well as tetragonal (CeCu_2Si_2) symmetry. To the best of our knowledge, however, only in the cubic case (UBe_{13}) have predictions regarding the temperature dependence of $\chi_s^{(s)}(\mathbf{Q},0)/\chi_s^{(n)}(\mathbf{Q},0)$ been made²⁴ for all p -wave superconducting states [$\chi_s^{(s)}(\mathbf{Q},0)$ and $\chi_s^{(n)}(\mathbf{Q},0)$ are the generalized spin susceptibilities in the superconducting and normal states, respectively]. We will therefore limit our discussion to the case of cubic symmetry (UBe_{13}). In this case (UBe_{13}) there are 12 possible p -wave superconducting states. It is interesting to notice that neither the polar nor ABM states, familiar from ^3He , are possible in a system with cubic symmetry. The generalized spin susceptibility²⁴ $\chi_s(\mathbf{Q},0)$ is temperature independent below T_c only for the

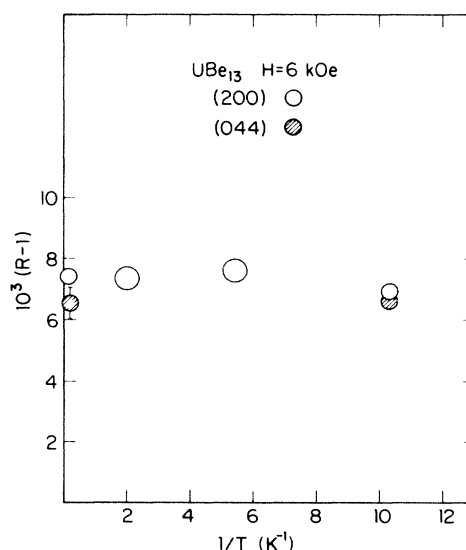


FIG. 4. Temperature dependence of the residual polarization ratio $(R - 1)$ of the (200) and (044) reflections of UBe_{13} for an applied field of 6 kOe. The size of the circles indicates the experimental precision.

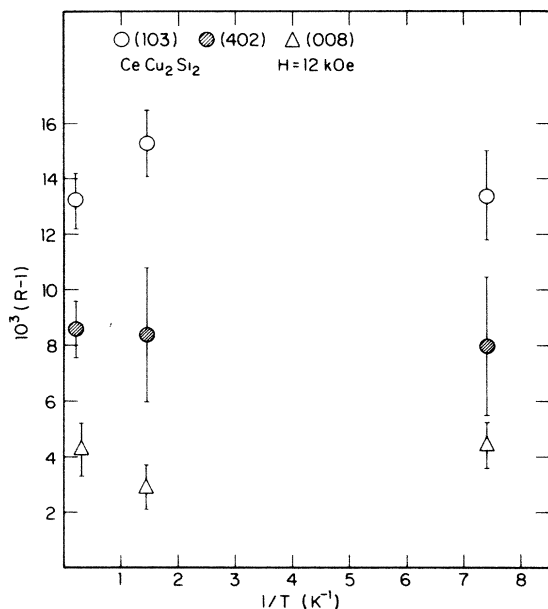


FIG. 5. Temperature dependence of the residual polarization ratio ($R - 1$) of the (103), (402), and (008) reflections of CeCu_2Si_2 for an applied field of 12 kOe.

states 4, 6, and 7 (identified by the serial numbers assigned to them²² by Blount). Thus, if $\chi_s(\mathbf{Q}, 0)$ is a significant part of $\chi(\mathbf{Q}, 0)$ in the normal Fermi-liquid state, our results suggest that one of these states is the superconducting state of the system. Actually, in a recent Letter Joynt,

Rice, and Ueda²⁵ proposed a phase diagram for the $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ system, which restricts the number of possible superconducting p states of UBe_{13} (phase A in the notation of Joynt *et al.*²⁵) from 12 to 5. From these five states, only state 4 exhibits²⁵ a temperature-independent $\chi_s(\mathbf{Q}, 0)$ and therefore could be considered as a good candidate for the superconducting state of UBe_{13} .

In summary, our experiments have shown that at 4.2 K, the induced magnetization in UPt_3 , UBe_{13} , and CeCu_2Si_2 is predominantly of f -electron character, and that $\chi(\mathbf{Q}, 0)$ for these materials is temperature independent below 4.2 K. We have illustrated for the case of UBe_{13} the kind of restrictions that the temperature dependence of $\chi_s(\mathbf{Q}, 0)$ can impose on the possible superconducting states of the heavy-electron systems. Clearly, quantitative theoretical calculations of $\chi_s(\mathbf{Q}, 0)$ and its temperature dependence below T_c for these systems would be a valuable guide in assessing the proper experimental precision of future experiments. In addition, a more detailed characterization of the purity of the samples used in the experiments is essential in assessing whether the effect on $\chi_s(\mathbf{Q}, 0)$ of spin-orbit scattering by impurities is significant. Measurements to assess this effect are certainly needed for the case of CeCu_2Si_2 , since Josephson tunneling experiments²⁶ strongly indicate s -wave pairing for this system.

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