Multiple-q Magnetic Structure in $CeAl_2$

S. M. Shapiro and E. Gurewitz[a]
Brookhaven National Laboratory, Upton, New York 11973

and

R. D. Parks and L. C. Kupferberg[b]
Department of Physics, University of Rochester, Rochester, New York 14627

(Received 25 April 1979)

New neutron-diffraction measurements on single crystals of $CeAl_2$ confirm the previously reported incommensurate antiferromagnetic ground state with peaks at $\vec{q}_I = (\frac{1}{2}\pm\alpha, \frac{1}{2}\mp\alpha, \frac{1}{2})$. In addition, weaker superlattice peaks are observed at $\vec{q}_c = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ and their position and temperature dependence can be explanined in terms of a multiple-q structure in $CeAl_2$. This appears to be the first definitive evidence for a multiple-q magnetic structure.

The rare-earth metals and compounds have served as invaluable proving grounds for testing many of the fundamental ideas of magnetism.[1] One of the least understood elements of this series is Ce. This is surprising since it contains only one f electron and therefore might be considered as the simplest of the rare-earth metals. The complicated behavior arises because of the large spatial extent of the $4f$ wave function of Ce, in contrast to the other rare-earth elements. This, in turn, leads to hybridization effects between the localized $4f$ electrons and the broadband s and d electrons which are manifested in (1) the Kondo effect in numerous dilute Ce systems[2]; (2) mixed valence in numerous concentrated systems (e.g., Ce-based fcc systems[3]); and (3) Kondo-like behavior in other concentrated systems (e.g., $CeAl_2$ and $CeAl_3$).[3]

In fact, $CeAl_2$ has been considered as a concentrated Kondo system with a high Kondo temperature ($T_K \sim 10$ K).[3] At lower temperatures ($T < 4.0$ K) the system was recently shown to order in an incommensurate antiferromagnetic structure.[4] The modulated moment in this system at low temperatures was postulated to be due to the strong singlet coupling between the f electron and the conduction electrons.[4]

We present below neutron-scattering studies performed on single crystals of $CeAl_2$ and show that the magnetic ground state of this system is a clear example of a multiple-q structure and differs conceptually from that originally proposed.[4] A triple-q structure was first proposed to explain the observations on the magnetic structure of Nd.[5] However, ambiguities remain in the experimental confirmation of this type of structure in Nd.[6] In the present experiment on $CeAl_2$, the position and the temperature dependence of newly observed peaks is consistent with a triple-q-type structure.

The single crystals were prepared by remelting boules of $CeAl_2$ in a sealed tantalum tube followed by a traveling-heat-zone solidification. The neutron-diffraction studies were performed on a triple-axis spectrometer at the Brookhaven National Laboratory high-flux-beam reactor. Most measurements were performed with an incident energy of $E_i = 13.5$ meV and collimation of 20′-40′-40′-40′ between reactor and monochromator, monochromator and sample, and sample analyzer and analyzer-detector, respectively. Pyrolytic graphite (PG) was used as a monochromator and analyzer in addition to the use of two 5-cm-long PG filters to eliminate any higher-order contamination. The intensity of several of the peaks was measured as a function of incident energy to determine whether multiple scattering contaminated the peaks. All peaks reported are free of multiple scattering.

The sample was oriented with the $[11\bar{2}]$ direction perpendicular to the scattering plane, which allowed for observation of the maximum number of satellites. The scattering plane is shown in Fig. 1. The allowed Bragg peaks are indicated by solid circles and the solid triangles show the incommensurate peaks previously reported. These peaks occur at $\vec{q}_I = (\frac{1}{2}\pm\alpha, \frac{1}{2}\mp\alpha, \frac{1}{2})$ when reduced to the first Brillouin zone. In our measurement, $\alpha = 0.108 \pm 0.001$, which is slightly less than the value 0.112 reported by Barbara *et al.*[4] and may be due to differences in the sample. According to Barbara *et al.*[4], the structure can be viewed as (111) planes of parallel spins pointing along the [111] direction. The spins in adjacent (111) planes point in the opposite direction. Superimposed upon this is an additional modulation of

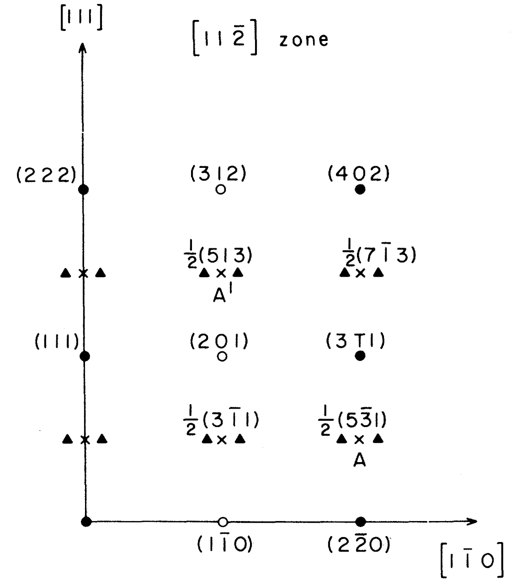


FIG. 1. The (11$\bar{2}$) scattering plane. Closed (open) circles represent allowed (not allowed) nuclear Bragg peaks of the *Fd*3*m* structure. The triangles (▲) are the incommensurate antiferromagnetic peaks at $q_i = (\frac{1}{2}\pm\alpha, \frac{1}{2}\mp\alpha, \frac{1}{2})$ with $\alpha = 0.018$. The crosses (×) are the new peaks appearing at $q_c = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.

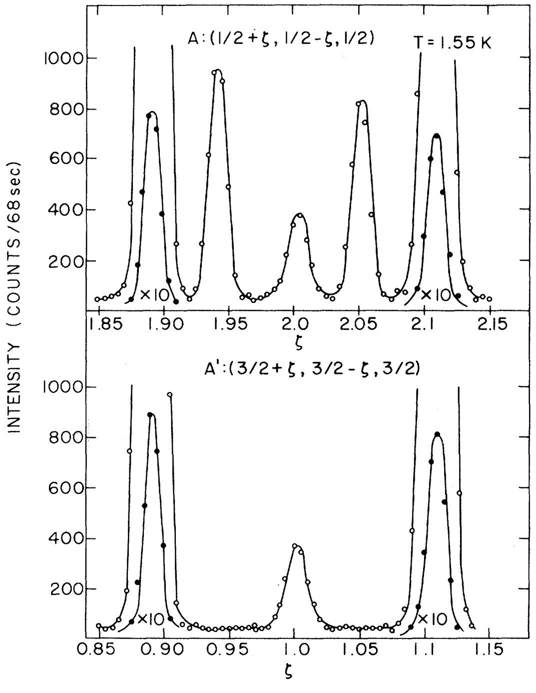


FIG. 2. $E = 0$ scans along the [1$\bar{1}$0] direction centered at point *A* and *A'* of Fig. 1.

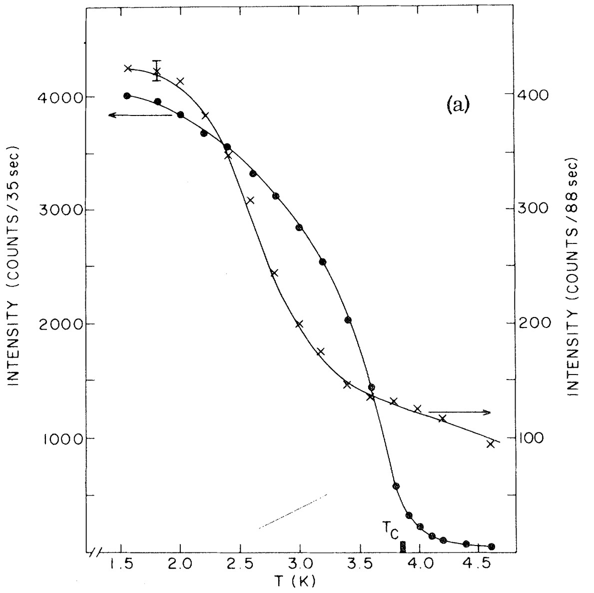
the amplitudes of the spins along a [1$\bar{1}$0] direction perpendicular to the [111] direction. This modulation, which exists down to very low temperatures has been considered as strong evidence of the Kondo coupling in this system.[4] Figure 2 shows q scans along the [1$\bar{1}$0] direction centered at *A* and *A'* of Fig. 1. The intense peaks at $\vec{q}_I = (\frac{1}{2}+\alpha, \frac{1}{2}-\alpha, \frac{1}{2})$ are due to the modulation, α, along (1$\bar{1}$0). The two peaks in curve *A* at $\alpha/2 = 0.054$ are due to modulations along equivalent [$\bar{1}$01] and [01$\bar{1}$] directions which make an angle of 60° with respect to the $\frac{1}{2}$(5$\bar{3}$1). Because of the finite vertical resolution we intersect part of this peak but its maxima lie above and below the scattering plane. The peaks are absent at *A'* since they make an angle with the scattering plane which is beyond the vertical resolution of the instrument. The new feature shown in both scans in the peak at $\vec{q}_c = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The temperature dependence of the peaks at $\vec{q}_I$ and $\vec{q}_c$ is shown in Fig. 3(a). The more-intense incommensurate peak exhibits a smooth variation of intensity with temperature. The transition appears to be second order with $T_N = 3.87 \pm 0.02$ K. The tail above T_N corresponds to critical scattering and line broadening has been observed. Using this T_N, we find a power-law behavior $I(q_I) \sim (T_N - T)^{2\beta}$, where β is near $\frac{1}{3}$. More detailed measurements are required to give precisely the exponent. The intensity of the weaker commensurate peak has a radically different type of behavior and seems to follow a power law with $2\beta > 1$.

The position of the commensurate peak and its temperature dependence can be understood from a simple Landau expansion of the free energy in terms of the magnetization:

$$F = F_0 + a(T)(M_{q_1}{}^2 + M_{q_2}{}^2 + M_{q_3}{}^2) + a' M_{q_c}{}^2 + \sum_{i=1}^{3} M_{q_i}{}^4 + c M_{q_1} M_{q_2} M_{q_3} M_{q_c}, \qquad (1)$$

where M_{q_i} ($i = 1, 2, 3$) are the q components of the magnetization corresponding to the three (1$\bar{1}$0) modulation directions perpendicular to [111], i.e., $\vec{q}_1 = (\frac{1}{2}+\alpha, \frac{1}{2}-\alpha, \frac{1}{2})$, $\vec{q}_2 = (\frac{1}{2}-\alpha, \frac{1}{2}, \frac{1}{2}+\alpha)$, and $\vec{q}_3 = (\frac{1}{2}, \frac{1}{2}+\alpha, \frac{1}{2}-\alpha)$. Because of the required translational invariance, only terms with $\sum_i \vec{q}_i = \vec{G}$, where $\vec{G}$ is a reciprocal-lattice vector or zero, are allowed. From the last term, this implies that $\vec{q}_c = \vec{q}_1 + \vec{q}_2 + \vec{q}_3$ or $\vec{q}_c = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ from the above definitions of q_i.

Also, from the last term in Eq. (1), the temperature dependence of M_{q_c} is determined by M_{q_i}. Since each M_{q_i} has the same temperature depen-



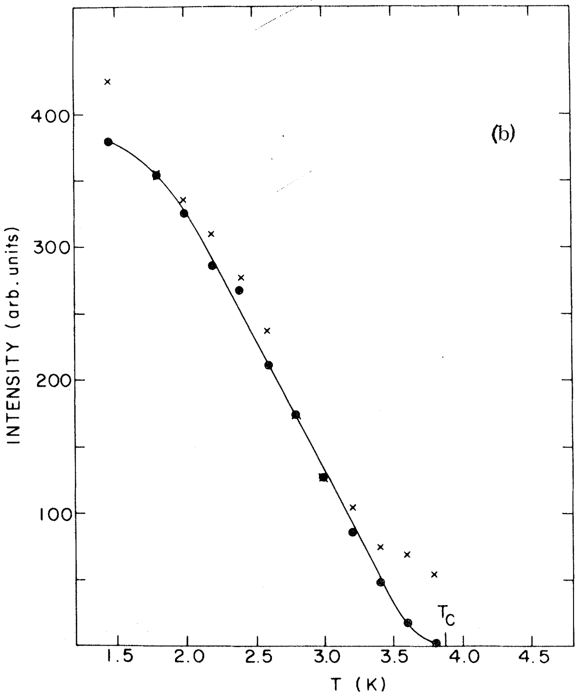


FIG. 3. (a) Temperature dependence of incommensurate peak measured at $(\frac{1}{2}+\zeta, \frac{1}{2}-\zeta, \frac{1}{2})$ with $\zeta = 1.892$ (open circles) and the commensurate peak measured at $\frac{1}{2}(5,\bar{3}, 1)$ (crosses). (b) $[I(\frac{1}{2}+\zeta, \frac{1}{2}-\zeta, \frac{1}{2})]^3$ with $\zeta = 1.892$ and $I(\frac{1}{2}(5,\bar{3},1))$ showing that these curves follow the same temperature dependence. The curves were normalized at 2.8 K.

dence, minimizing F with respect to M_{q_c} and neglecting fluctuations

$$M_{q_c}(T) \propto M_{q_i}^{3}(T).$$

This behavior is consistent with our measurements as shown in Fig. 3(b) where we plot $I^3(\vec{q})$ and $I(\vec{q}_c)$ on the same scale. The intensities were normalized at $T = 2.8$ K. The curves coincide except for temperature near T_c where the intensity of $I(q_c)$ is weak such that background subtraction limits the accuracy. A more detailed study of the critical properties of $CeAl_2$ is in preparation.

Thus, it appears that the magnetic structure of $CeAl_2$ is a triple-q structure for each [111] domain. Bak and Lebech[5] recently proposed a triple-q structure to explain the experimental observation in the ordered state of hexagonal Nd. This was the first and, until the present work, the only reported evidence for a multiple-q magnetic structure. In order to explain the existence of certain peaks, they assumed a coupling of the magnetization to the lattice and the unknown peaks were reported to be nuclear in origin. However, recent polarized-neutron-beam and x-ray experiments have shown that these peaks are magnetic in origin, not nuclear,[6] so that the experimental verification of a triple-q structure in Nd is incomplete. In the present case, no coupling to the lattice is necessary and all peaks are considered as magnetic in origin. A polarized-neutron-beam or x-ray experiment would be useful to confirm this.

In the above treatment, we considered $CeAl_2$ as a triple-q structure with the three wave vectors $\vec{q}_1$, $\vec{q}_2$, $\vec{q}_3$ being perpendicular to the [111] direction. However, there are eight equivalent $\langle 111\rangle$ directions and associated with each one are three perpendicular vectors corresponding to the $\langle 111\rangle$ directions. Thus, to be exact, $CeAl_2$ should be discussed in terms of a 24-q structure consisting of eight triple-q structures.

The magnetic structure in real space is the sum of the individual q components M_{q_i} so that

$$M(\vec{r}) = \sum_{i=1}^{24} M_{q_i} \cos(\vec{q}_i \cdot \vec{r} + \varphi),$$

where φ is an arbitrary phase. This is not easily visualized in real space and the magnitude of the spins varies greatly depending upon the direction. Importantly, the magnetic moment is still modulated and the idea of a Kondo coupling of the f electron to the conduction electrons as proposed by Barbara *et al.*[4] can equally well apply to the triple-q structure.

We gratefully acknowledge our discussions with Per Bak, B. Barbara, W. C. Koehler, D. E. Moncton, R. M. Moon, and S. K. Sinha. This research was supported in part by the Division of

Basic Energy Sciences, U. S. Department of Energy, under Contract No. EY-76-C-02-0016, and in part by the U. S. Office of Naval Research and the National Science Foundation.

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[a]Permanent address: Nuclear Research Centre-Negev, P. O. B. 9001, Beer-Sheva 84190, Israel.

[b]Present address: Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139.

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Detection of EPR Transitions of Muonium in Quartz by Muon-Spin Rotation

J. A. Brown, R. H. Heffner, and M. Leon
Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87545

and

S. A. Dodds and D. A. Vanderwater
Rice University, Houston, Texas 77001

and

T. L. Estle
Rice University, Houston, Texas 77001, and Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87545

(Received 23 July 1979)

Magnetic resonance transitions of muonium in quartz have been detected by their effect on muon-spin-rotation frequency spectra. This technique can be used to observe EPR transitions which are not directly observable in the normal muon-spin-rotation spectrum. The coherence effects of an intense rf magnetic field on two strongly coupled spins are decribed theoretically and are found to explain the data in detail.

Positive-muon-spin-rotation experiments normally involve the study of the time history of the free precession of the muon spin by observation of the decay positrons.[1] For positive muons stopped in many insulators and semiconductors, an appreciable fraction of the muons bind an electron forming a muoniumlike state. In the case of quartz, a state very similar to muonium in vacuum is formed, having a reported hyperfine frequency of 4463 ± 300 MHz with a very slight anisotropy[2] (which can be ignored in the present work). Two frequency components are measured in the muonium time-differential muon-spin-rotation data, corresponding to the two allowed magnetic-dipole transitions within the $F=1$ triplet.

In this Letter we report for the first time the effect of EPR transitions on the muon-spin-rotation spectrum. We present muon-spin-rotation data taken while a single-crystal sample of quartz was in an intense rf magnetic field with a frequency near one of the two observed muonium frequencies. The resulting data give a very complete picture of the coherent response of two strongly coupled spins to an intense rf magnetic field—in effect, a double electron-muon resonance (DEMUR). The information obtained is similar to, but more detailed than, that obtained by various other double-resonance techniques (electron-nuclear double resonance, inequivalent-nuclear double resonance, electron double resonance, optically detected magnetic resonance).[3] In particular one observes the precessional component at the radio frequency and its two side bands, none of which can be probed in standard double-resonance experiments. A theoretical description of these effects is outlined and is found to be in excellent agreement with the data. Finally, it is argued that the DEMUR technique