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Multiple-$\vec{q}$ Structure or Coexistence of Different Magnetic Phases in $CeAl_2$?

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Neutron diffraction experiments under perturbations, such as uniaxial stress and hydrostatic pressure, suggest strongly that $CeAl_2$ exhibits two magnetic phases: a single-$\vec{q}$ collinear modulated structure and a type-II antiferromagnetic structure.

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The intermetallic Laves-phase compound $CeAl_2$ exhibits sinusoidally modulated magnetic structure,[1,2] which implies a moment reduction attributed to a Kondo-like coupling of the cerium 4f electrons to the conduction band. This structure develops with a propagation vector $\vec{q}_i = (\frac{1}{2}+\tau, \frac{1}{2}-\tau, \frac{1}{2})$; τ is found to be temperature independent and its value is $\tau = 0.110$ ($\pm$ 0.002).[2] The Fourier component of the magnetization $\vec{m}_{\vec{q}_i}$ is parallel to the [111] direction. Because of the symmetry of paramagnetic space group ($Fd3m$), there are twenty-four nonequivalent $\vec{q}_i$ vectors in the Brillouin zone (see Fig. 1) which leads to the coexistence of twenty-four Fourier components $\vec{m}_{\vec{q}_i}$. Orig-

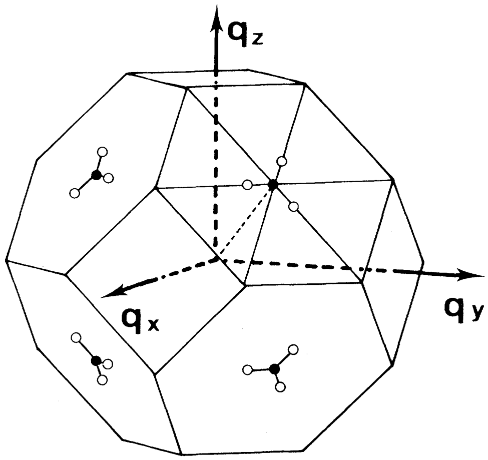

FIG. 1. Brillouin zone of the fcc lattice. Full circles represent L points at $\vec{q}_c = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ positions. Open circles correspond to $\vec{q}_i = (\frac{1}{2}+\tau, \frac{1}{2}-\tau, \frac{1}{2})$ positions.

inally, the magnetic structure of $CeAl_2$ was discussed in terms of independent $\vec{q}_i$ vectors; the net result is a single-$\vec{q}$ collinear structure, the overall cubic symmetry being preserved by the coexistence of twenty-four magnetic domains. However, in a recent Letter[3] a multiple-$\vec{q}$ structure has been proposed for $CeAl_2$. Evidence for a coupling of different $\vec{q}$ vectors was the observation of weak superlattice peaks corresponding to a propagation vector $\vec{q}_c = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. In this picture, to preserve the translational invariance of the free energy, three $\vec{q}_i$ vectors, namely $\vec{q}_1 = (\frac{1}{2}+\tau, \frac{1}{2}-\tau, \frac{1}{2})$, $\vec{q}_2 = (\frac{1}{2}-\tau, \frac{1}{2}, \frac{1}{2}+\tau)$, $\vec{q}_3 = (\frac{1}{2}, \frac{1}{2}+\tau, \frac{1}{2}-\tau)$, would couple to give intensity at $\vec{q}_c = \vec{q}_1 + \vec{q}_2 + \vec{q}_3$ position with an intensity such as $\vec{m}_{\vec{q}_c}(T) \sim \vec{m}_{\vec{q}_i}{}^3(T)$. All the experimental data presented in the work by Shapiro *et al.*[3] suit this model. Again this work raises the problem of the coupling between different wave vectors among other magnetic materials[4]; neodymium had been thought to exhibit a triple-$\vec{q}$ structure[5] until subsequent studies[6,7] showed that the triple-$\vec{q}$ structure was not the correct model of neodymium.

In the case of $CeAl_2$, there are experimental evidences which show that the superlattice peaks at $\vec{q}_c$ are not due to a coupling of different $\vec{q}_i$ but rather to the existence of another magnetic structure of $CeAl_2$. Our studies were stimulated by the observation of commensurate peaks at $\vec{q}_c$ in diluted systems such as $Ce_{1-x}Pr_xAl_2$.[8] As a magnetic origin is invoked by the two explanations, a study of neutron diffraction with use of polarization analysis would not give a suitable answer as in the neodymium case. Consequently, we submitted $CeAl_2$ single crystals to small uniaxial stress in order to decouple nonequivalent propagation vectors. Our measurements show that the three $\vec{q}_i$ vectors around $\vec{q}_c$ are not coupled. Further measurements demonstrate that hydrostatic pressure induces a simple type-II antiferromagnetic structure, which is characterized by superlattice intensity showing up at $\vec{q}_c$ positions. The two magnetic structures of $CeAl_2$ coexist over a wide range in pressure; the large pressure hysteresis that we observed accounts for the existence of traces of the type-II structure at room pressure.

The single crystals were prepared by the Bridgman technique. Tungsten crucibles have been used; indeed as shown by the studies of the isomorphous compound $NdAl_2$,[9] only tungsten crucibles yield noncontaminated samples. (In a crystal grown in a tantalum crucible, the amount of Ta ranges from 1.2% to 3%, whereas the amount of tungsten is only 120 ppm with use of tungsten crucible.) Two samples have been cut from the ingot: a parallelepiped with the faces perpendicular to [111], [$1\bar{1}0$], and [$11\bar{2}$] for the measurements under stresses, and a cylinder with axis parallel to [$11\bar{2}$] for hydrostatic pressures.

Hydrostatic pressure was applied with use of a standard alumina pressure cell[10]; deuterated methanol-ethanol mixture was used as a pressure medium. Uniaxial stress was applied perpendicular to the scattering plane; the load was transmitted to the sample through two hardened steel pistons which were coated with teflon to reduce friction on the sample faces. The actual applied load was determined from the output of a strain gauge bridge which measured deformations of the uniaxial stress assembly at the sample position.

Hydrostatic-pressure cell or uniaxial-stress assembly was mounted within a standard cryostat. The neutron diffraction experiments were carried out on the D1A two-axes spectrometer at the Institut Laue-Langevin. A focusing Ge(311) monochromator was used at a wavelength of 1.91 Å; as discussed below the vertical divergence of the beam (6°) allowed the observation of the satellites which were not exactly in the scattering plane.

The first Brillouin zone for a fcc lattice is shown in Fig. 1. The open circles correspond to nonequivalent $\vec{q}_i$ vectors. Different Fourier components $\vec{m}_{\vec{q}_i}$ correspond to these twenty-four $\vec{q}_i$ vectors. In an unperturbed crystal the $\vec{m}_{\vec{q}_i}$ are equally distributed. An applied uniaxial stress will differentiate some $\vec{m}_{\vec{q}_i}$. If the actual magnetic structure was a multiple-$\vec{q}$ structure, then

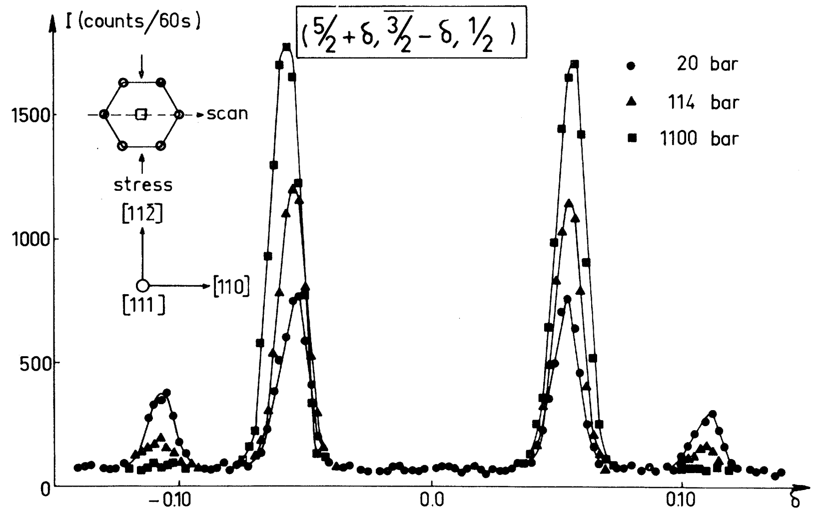

FIG. 2. Scans along the [1$\bar{1}$0] directions around a $\vec{q}_c$ position at different values of the applied stress. The inset shows the positions of the magnetic peaks around $\vec{q}_c$ with respect to the direction of uniaxial stress.

the $\vec{m}_{\vec{q}_i}$, which would be coupled at zero stress, would remain coupled at low applied stress. In particular, in the case of triple-$\vec{q}$ structure, the relative intensities of the three magnetic peaks at $\vec{q}_i$ around each $\vec{q}_c$ position would not depend on the applied stress. On the opposite if there is no coupling of the $\vec{m}_{\vec{q}_i}$, then these relative intensities will vary, provided that the applied stress differentiates the $\vec{m}_{\vec{q}_i}$.

We have applied uniaxial stress along [11$\bar{2}$]; magnetic intensities have been observed in different Brillouin zones for two directions of the moments [111] and [11$\bar{1}$] around $\vec{q}_c=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$, and $\vec{q}_c(\frac{1}{2},\frac{1}{2},-\frac{1}{2})$. In the reciprocal space, the magnetic satellites are on a hexagon (see Fig. 2) formed by two equilateral triangles coming from two adjacent Brillouin zones. Scans in reciprocal space were perpendicular to the [11$\bar{2}$] stress. Scans performed along the [1$\bar{1}$0] direction, around the point $(\frac{5}{2},\frac{3}{2},\frac{1}{2})$, are shown in Fig. 2, for three different values of the applied stress. At $\sigma=20$ bars, we observe four magnetic reflections: two of them ($\delta=0.109$) originate from the two extremities of the hexagon; they are in the scattering plane. The other two reflections are associated with the four points of the hexagon which are not in the scattering plane. They are separated by $\Delta\delta=0.109$ which corresponds effectively to $\tau/2$. Because of the large vertical divergence of the beam, almost the whole intensity of such reflection could be measured. At zero stress their intensity is approximately twice that of the magnetic reflection at $\pm\tau$, because two reflections are superimposed. At 114 bars the peaks at $\pm\tau/2$ increase whereas those at $\pm\tau$ decrease strongly; those peaks are totally suppressed at higher stress. Comparisons of intensities observed in different zones show that Fourier components $\vec{m}_{\vec{q}_i}$ with $\vec{q}_i$ parallel to the stress are favored with increasing stress. This very rapid variation of the relative intensities shows that the $\vec{m}_{\vec{q}_i}$ are not coupled; any combination of three $\vec{q}$ vectors out of the twelve vectors can be ruled out. It might be argued that applied stress would destroy any eventual coupling; however, the elastic energy per Ce atom can be estimated from the elastic constants measured[11] and is found to be 0.4 K at 1 kbar, which is much less than $T_N=3.8$ K. The relative intensities are very sensitive to low applied stress: Even at 114 bars important changes are observed. This means that our observations are not due to a breaking of the cubic symmetry.

In the scan presented in Fig. 2, it was not possible to observe any scattering with $\vec{q}_c=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$. However, a peak at $\vec{q}_c$ was clearly observed by Shapiro *et al.*[3]; careful investigation of other samples have also indicated the existence of such reflections but with a smaller relative intensity.[2,8] Mixed $(Ce_{1-x}Pr_x)Al_2$ polycrystalline samples exhibit (besides normal $CeAl_2$ magnetic lines) a well-defined $\vec{q}_c$ magnetic peak, whose intensity depends on the amount of Pr impurities.[8] This peak is characteristic of a type-II antiferromag-

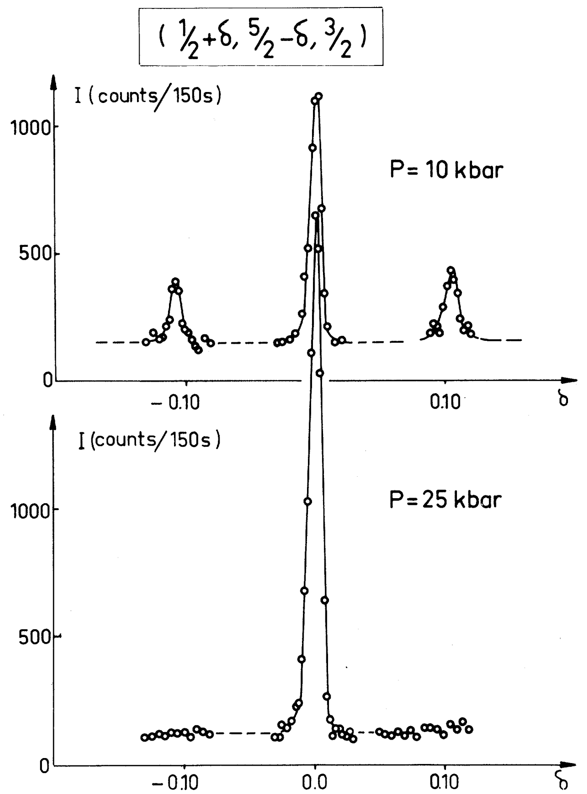

FIG. 3. Scans along the [1$\bar{1}$0] direction around a $\vec{q}_c$ position at two values of the hydrostatic pressure. At 25 kbar, only the commensurate peak at $\vec{q}_c$ can be observed.

netic structure, which coexists with the modulated structure: The impurities can be considered as responsible for the pinning of the modulation.

Hydrostatic pressure has been used to modify the relative strength of the interactions. At 25 kbar, we have found that all the reflections at $\vec{q}_i$ have disappeared, whereas strong intensity can be observed at $\vec{q}_c$ positions (Fig. 3). Those lines are characteristic of a simple type-II antiferromagnetic and have been measured in different Brillouin zones.[12] When releasing the pressure coexistence of the two phases is observed (Fig. 3). The large pressure hysteresis can explain the existence at room pressure of residues of the high-pressure phase which are stabilized by point defects or impurities. As it can be seen in Fig. 4, the simple type-II antiferromagnetic phase and the sinusoidally modulated phase have different Néel temperatures. Thermal variations of the $\vec{q}_i$ and $\vec{q}_c$ satellites which have been measured in the coexistence region show clearly different Néel temperatures. A $P = 10$ kbar the $\vec{q}_i$ satellites with $T_{N_i} = 3.3$ K follow a power law with $\beta_i \simeq 0.5$. The $\vec{q}_c$ satellites have $\beta_c \sim 0.5$ with $T_{N_c} = 2.7$ K. This

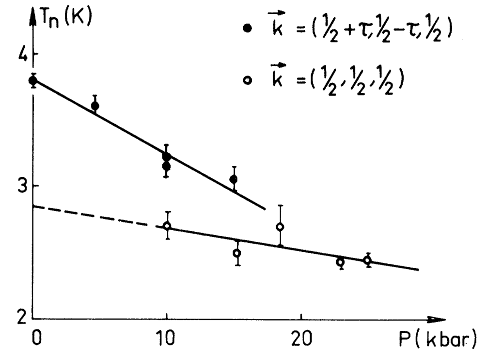

FIG. 4. Pressure dependence of the Néel temperature of the sinusoidally modulated phase (full points), and type II antiferromagnetic phase (open points).

difference in Néel temperatures might be a reason for the differences in temperature dependence of the intensities reported in Ref. 3.

We settle on an alternative magnetic structure of $CeAl_2$—other than that proposed by Shapiro *et al.*—a single-$\vec{q}$ collinear structure. The weak intensity that can be observed at commensurate $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ positions reveals the existence of traces of a simple type-II antiferromagnetic structure, whose internal energy is not very different from that of the sinusoidally modulated structure.

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