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Author(s): M. Kenselmann, T. Strassle, C. Niedermayer, M. Sigrist, B. Padmanabhan, M. Zolliker, A.D. Bianchi, R. Movshovich, E.D. Bauer, J.L. Sarrao, J.D. Thompson

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Coupled Superconducting and Magnetic Order in $CeCoIn_5$

M. Kenzelmann[1,2], Th. Strässle[3], C. Niedermayer[3], M. Sigrist[4], B. Padmanabhan[3], M. Zolliker[1], A. D. Bianchi[5], R. Movshovich[6], E. D. Bauer[6], J. L. Sarrao[6] and J. D. Thompson[6]

(1) Laboratory for Developments and Methods, Paul Scherrer Institute, CH-5232 Villigen, Switzerland

(2) Laboratory for Solid State Physics, ETH Zurich, CH-8093 Zurich, Switzerland

(3) Laboratory for Neutron Scattering, ETH Zurich & Paul Scherrer Institute, CH-5232 Villigen, Switzerland

(4) Institut für Theoretische Physik, ETH Zurich, CH-8093 Zurich, Switzerland

(5) Département de Physique et RQMP, Université de Montréal, Montréal, QC H3C 3J7, Canada.

(6) Condensed Matter and Thermal Physics, Los Alamos National Laboratory, Los Alamos, NM 87545, USA.

Strong magnetic fluctuations can provide a coupling mechanism for electrons that leads to unconventional superconductivity. Magnetic order and superconductivity have been found to coexist in a number of magnetically-mediated superconductors, but these order parameters generally compete. We report that close to the upper critical field, $CeCoIn_5$ adopts a multi-component ground state that simultaneously carries cooperating magnetic and superconducting order. Suppressing superconductivity in a first-order transition at the upper critical field leads to the

simultaneous collapse of the magnetic order, showing that superconductivity is necessary for the magnetic order. A symmetry analysis of the coupling between the magnetic order and the superconducting gap function suggests a form of superconductivity that is associated with a non-vanishing momentum.

$CeCoIn_5$ is a clean ambient-pressure d-wave superconductor (*1*) and crystallizes in a tetragonal structure (Fig. 1). Due to its proximity to a magnetic quantum critical point, it features strong antiferromagnetic correlations that result in an enhancement of the effective electronic mass and heavy-fermion behaviour (*2*). However, $CeCoIn_5$ undergoes a transition to superconductivity before magnetic order can be established (*1*, *2*). The superconducting gap function has a $d_{x^2-y^2}$ symmetry (*3-6*), and it is generally believed that superconductivity in $CeCoIn_5$ is mediated by magnetic fluctuations. The Fermi surface is strongly two-dimensional (*7, 8*) and superconductivity in an applied field is Pauli-limited (*9*), i.e. it is destroyed by a coupling of external magnetic fields to the spins of the Cooper pairs and not by orbital depairing. $CeCoIn_5$ features an unusual field-temperature (H-T) phase diagram (*10, 11*): below $T_0 = 0.31 T_c = 1.1$ K, the transition from the normal to the superconducting state is first order (*9, 10*). Further, there is evidence for a second superconducting phase, the "**Q** phase" which exists only for $T < 0.3$ K and high fields close to the upper critical field (*10, 12, 13*). It has been suggested (*10, 14*) that this high-field phase may represent a superconducting phase that was proposed by Fulde, Ferrell, Larkin and Ovchinnikov (FFLO) and that carries a finite momentum q as a result of the Zeeman splitting of the electron bands (*15, 16*).

A rich interplay between magnetic order and superconductivity is characteristic of heavy-fermion superconductors, with either magnetic order preceeding the onset of superconductivity or superconductivity occurring in the vicinity of a quantum critical point (*17, 18*). Superconductivity in $CeCoIn_5$ is special in that it occurs close to a magnetic quantum critical point, but so far there has been no direct evidence of long-range magnetic order anywhere in the H-T phase diagram (*2*). However, there is microscopic evidence from NMR measurements for field-induced magnetism for high fields ($\sim H = 11$ T) in the tetragonal plane and for temperatures below which the $H_{c2}(T)$ phase boundary becomes first order (*19*). The NMR results were interpreted as evidence that the **Q** phase is a phase where superconductivity and magnetic order co-exist, but the character of the superconducting state could not be ascertained.

We have used high-field neutron diffraction to directly search for magnetic Bragg peaks within the **Q** phase. The measurements were done at low temperatures and the field was applied along the crystallographic [1 -1 0] direction in the tetragonal basal plane. For this field direction, the upper critical field in the zero-temperature limit is $H_{c2}(0)$= 11.4 T. Fig. 2 shows the neutron diffraction data for wave-vectors along the (h, h, 0.5) reciprocal direction. For $10.5 \text{ T} < H < 11.4 \text{ T}$, neutron scattering provides clear evidence of Bragg peaks that arise from a magnetic structure that is modulated with the ordering wave-vector $\mathbf{Q} = (q, q, 0.5)$ (*20*) and that are present at neither higher nor lower fields outside the **Q** phase (hence its name). The width of the peaks is resolution limited, so the magnetic order extends over a length scale of $\xi > 60$ nm. This is much larger than the diameter of vortex cores, which is of the order of the coherence length $\xi_0 \sim 10$ nm (*5*), and so magnetic order is not limited to the vortex cores.

Fig. 3 shows the field and temperature dependence of the peak intensity of the $\mathbf{Q} = (q, q, 0.5)$ magnetic Bragg peak obtained from a fit to a Gaussian line shape. The magnetic order at T = 60 mK has a gradual onset with increasing field and collapses at the superconducting phase boundary H_{c2} in a first order transition (Fig. 3A). The intensity of the magnetic Bragg peak can also be suppressed by increasing the temperature (Fig. 3B), the signal disappears at the same temperature where specific heat measurements show evidence of a second order phase transition (*10*). The neutron data suggest a transition that is second order in temperature but first order in field. The incommensuration q of the Bragg peak position is not field dependent, as can be seen in the inset of Fig. 3A. The H-T phase diagram (Fig. 1) shows that magnetic order exists only in the superconducting **Q** phase, and not in the normal phase, demonstrating that superconductivity is essential for magnetic order. Our results provide evidence that the ground state in this field and temperature range in the vicinity of $H_{c2}(0)$ has a multi-component order parameter which directly couples superconductivity and magnetism. This type of order is at least partly due to strong antiferromagnetic fluctuations arising from the proximity to a magnetic quantum critical point in $CeCoIn_5$.

Our experiment shows that the magnetic structure is a transverse amplitude-modulated incommensurate spin-density wave with the magnetic moments orientated along the tetragonal c axis, modulated with the incommensurate wave-vector (q, q, 0.5) perpendicular to the magnetic field. Neighbouring Ce^{3+} magnetic moments that are separated by a unit cell lattice translation along the c axis are anti-parallel (Fig. 1). The amplitude of the magnetic moment at $T = 60$ mK and $H = 11$ T of $m = 0.15(5)$ μ_B is considerably smaller than expected for the Ce^{3+} free ion, possibly due to the Kondo effect. The direction of the ordered magnetic moment is consistent with magnetic susceptibility measurements (*1*) that identify the c axis as the easy axis, and it is also consistent with zero-field inelastic neutron measurements in which strong antiferromagnetic fluctuations have been observed that are polarized along the c axis (*21*).

The magnetic structure that satisfies NMR data (*19*) was described by an ordering wave vector **Q** = (q, 0.5, 0.5) with unspecified q with the ordered magnetic moment along the applied field that was along the [100] direction. Our neutron measurements for field along the [1-10] direction reveal a magnetic order for which both the ordering wave-vector **Q** = (q, q, 0.5) and the ordered moments are perpendicular to the applied magnetic field – in contrast to the NMR data. This difference suggests that the direction of the incommensurate modulation, **Q**, depends on the field direction, and that the order wave-vector can be tuned with a rotation of the magnetic field in the basal plane. Finally, we point out that the absence of magnetic Bragg peaks at $H = 11$T for $T > 0.3$ K confirms the interpretation of the NMR measurements (*19*) that the fluctuations for 0.3 K $< T < T_0$ are short-ranged, and possibly only present inside the vortex cores.

The observation that magnetism exists only in the presence of superconductivity is in stark contrast to other materials where long-range magnetic order and superconductivity merely coexist for a small magnetic field or pressure range due to their different origins (*17, 18*). As no magnetic order is observed in $CeCoIn_5$ above the upper critical field H_{c2}, the relation between magnetic order and superconductivity is fundamentally different, and cannot be seen as a competition. Instead, it appears that $CeCoIn_5$ in fields greater than H_{c2} gives rise to strong antiferromagnetic fluctuations that condense into magnetic order with decreasing magnetic field only through the opening of an electronic gap and restructuring of the Fermi surface at the superconducting phase boundary. This means that the second order magnetic quantum phase transition is inaccessible, because in its proximity there is no energy scale associated with the antiferromagnetic fluctuations, and the superconducting energy gap becomes the dominant energy scale and determines the magnetic ground state properties.

The intimate link between superconductivity and magnetic order in $CeCoIn_5$ suggests the presence of a specific coupling between these order parameters (*22*). The multi-component magneto-superconducting phase can be reached via two second order phase transitions through a suitable path in the H-T phase diagram, justifying the construction of a phenomenological Landau coupling theory. Assuming the superconducting gap at zero field, Δ_d, is of $d_{x^2-y^2}$ symmetry, the possible coupling terms for magnetic fields in the basal plane, that preserve time reversal symmetry and conserves momentum can be written as $V_1 = \Delta^*_d M_q (H_x\Delta^{(5)}_{y,-q} + H_y\Delta^{(5)}_{x,-q}) + c.c.$, $V_2 =$

$\Delta^*_d M_q (H_x D_x - H_y D_y) \Delta^{(2)}_{-q} + c.c.$ and $V_3 = \Delta^*_d M_q (H_x D_y - H_y D_x) \Delta^{(3)}_{-q} + c.c.$ Here $(\Delta^{(5)}_{x,-q}, \Delta^{(5)}_{y,-q})$ belongs to the two-component even-parity Γ^+_5 state, $\Delta^{(2)}_{-q}$ and $\Delta^{(3)}_{-q}$ are the Γ^-_2 and Γ^-_3 odd-parity states (*23*) and M_q is the magnetic order parameter. Note that these additional superconducting order parameters include a finite momentum $-$q . (D_x, D_y) is the gauge invariant gradient. Introducing the magnetic field allows to couple M_q in linear order to preserve time reversal symmetry. These combinations allow for a second order phase transition within the superconducting phase and a first order transition to the non-magnetic normal state. For the coupling term V_2, no magnetic structure is induced for fields $\mathbf{H} \parallel [100]$. Given the weak dependence of the **Q** phase on the magnetic field orientation in the basal plane, our measurements suggest the presence of a V_1 or V_3 coupling term, inducing the finite-momentum even-parity Γ^+_5-state or the odd-parity Γ^-_3-state.

This Landau theory shows that incommensurate magnetic order induces a superconducting gap function that carries a finite momentum – the first experimental evidence of a superconducting condensate that carries a momentum. However, we show that this state may not arise purely from Pauli paramagnetic effects and the formation of a new pairing state between exchange-split parts of the Fermi surface, a state commonly known as the FFLO state (*15, 16*). In the FFLO state, the pairing state carries a momentum of the Cooper pair that depends on the magnetic field via $|q| = 2\mu_B H/\hbar v_F$, where v_F is the Fermi velocity. However, the inset of Fig. 3A shows that |q| is field independent in $CeCoIn_5$, at odds with this prediction, indicating that an additional superconducting pairing channel with finite momentum is induced in conjunction with the cooperative appearance of magnetic order.

A superconducting order that carries momentum illustrates the wealth of quantum phases that can exist in solid matter. The important microscopic role of magnetic fluctuations in the formation of Cooper pairs in $CeCoIn_5$ is self evident because superconductivity emerges at $H_{c2}(0)$ simultaneously with ordered magnetism.

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Figure 1: *H-T* phase diagram of $CeCoIn_5$ with the magnetically ordered phase indicated by the red shaded area. The blue and open circles indicate a first and second order transition measured by specific heat (*10*), respectively, separating the superconducting from the normal phase. The green circles indicate a second order phase transition inside the superconducting phase (*10*), and the red circles indicate the onset of magnetic order as measured in our experiment, showing that the magnetic order only exists in the **Q** phase. **Inset:** Magnetic structure of $CeCoIn_5$ at T = 60 mK and H = 11 T. The red arrows show the direction of the static magnetic moments located on Ce^{3+}, while the yellow and blue circles indicate the position of the In and Co ions. The solid red line indicates the amplitude of the Ce^{3+} magnetic moment along the c-axis, projected on the (*hhl*) plane.

Figure 2: The solid circles represent the neutron-scattering intensity at T = 60 mK for wave-vectors (*h*, *h*, 0.5) as a function of *h* for different fields as observed in the centre channel of the psd, showing the presence of a magnetic neutron diffraction peak at (1 - *q*, 1 - *q*, 0.5) with q = 0.44: **A** for *H* = 10.6 T, **B** *H* = 10.8 T, **C** *H* = 11 T and **D** *H* = 11.3 T. The grey circles in **A**,**B** represent the best estimate of the background, while in **C** they represent the neutron scattering intensity at *H* = 11 T and *T* = 400 mK and in **D** at *H* = 11.4 T and *T* = 60 mK. The solid lines in **A-D** are fits of a Gaussian function to the magnetic scattering.

Figure 3: Neutron-scattering intensity at (*q*, *q*, 0.5), **A** as a function of field at T = 60 mK , and **B** as a function of temperature at *H* = 11 T. The grey circles

represent the background scattering taken from the two nearest to the centre channels of the psd. The dashed line in **A** is a guide to the eye, while the dashed line in **B** describes the background and the onset of the magnetic order in a second order phase transition with $\beta = 0.365$ fixed to the critical exponent of the three-dimensional Heisenberg universality class. The **inset** shows that the *q* is field independent.

Supporting Online Material for

Coupled Superconducting and Magnetic Order in $CeCoIn_5$

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*To whom correspondence should be addressed. E-mail: michel.kenzelmann@psi.ch

This PDF file includes:

Materials and Methods

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Single-crystal samples of $CeCoIn_5$ were grown from In flux (*S1*). Neutron measurements were carried out using the RITA-II instrument at PSI and a superconducting magnet with a dilution refrigerator. The magnetic field produced by a superconducting magnet with a maximum field of 14.9T was applied in the tetragonal *ab*-plane along the [1-10] direction. No difference between field and zero-field cooling was detected in our measurements. All neutron measurements were performed with an incident energy of $E_i = 5$ meV and a position sensitive detector (psd) used in a multi-analyzer and monochromatic wave-vector dispersive imaging mode (*S2*). We used a Be filter before the analyzer in order to remove higher order neutrons, and the collimation around the sample was defined by the distance of the sample from the monochromator and analyzer (about 1.2 and 1.4 degree, respectively). The center channel of this psd setup was used to measure magnetic scattering, while the two adjacent channels were used to simultaneously measure the background.

Magnetic Bragg peaks were observed at wave-vectors $\mathbf{K}_m = \mathbf{K}_n \pm (q, q, 0.5)$ with $q = 0.44$. Here $\mathbf{K}_m$ and $\mathbf{K}_n$ indicate the magnetic and nuclear Bragg peak position and $\mathbf{Q} = (q, q, 0.5)$ is the magnetic ordering wave-vector that describes the magnetic modulation in the **Q** phase. Group theory was used to determine all allowed magnetic structures. The group of symmetry elements that leave **Q** invariant consists of four elements and is generated by powers of two mirror planes that leave **Q** invariant.

The magnetic structure at $H = 11$ T was determined by measuring a set of 8 magnetic Bragg peaks and comparing their relative intensities to those calculated for symmetry allowed structures. The magnetic form factor of Ce^{3+} was taken from calculations by Blume et al. (*S3*). There are three possible irreducible representations that describe different types of magnetic order: Two of them have the magnetic moments in the basal plane and are clearly excluded by the experiment ($\chi^2 > 30$). The third representation has the spin moments aligned along the *c*-axis and gives a good description of the magnetic order in $\mathrm{CeCoIn_5}$. The agreement for this model of the magnetic structure and the data is given by χ^2=8.76 and a R-factor R=0.37. Here χ^2 is the mean squared deviation between model and data in units of the variance and $R=(1/N)\ \Sigma_i|I^o{}_i - I^c{}_i|\ /\ I^o{}_i$ ($I^o{}_i$ and $I^c{}_i$ are the observed and calculated intensities, respectively). It is possible that the magnetic structure is a square-wave spin density wave, but our experiment does not have the sensitivity to observe weak higher order Bragg peaks that would provide evidence for such a structure.

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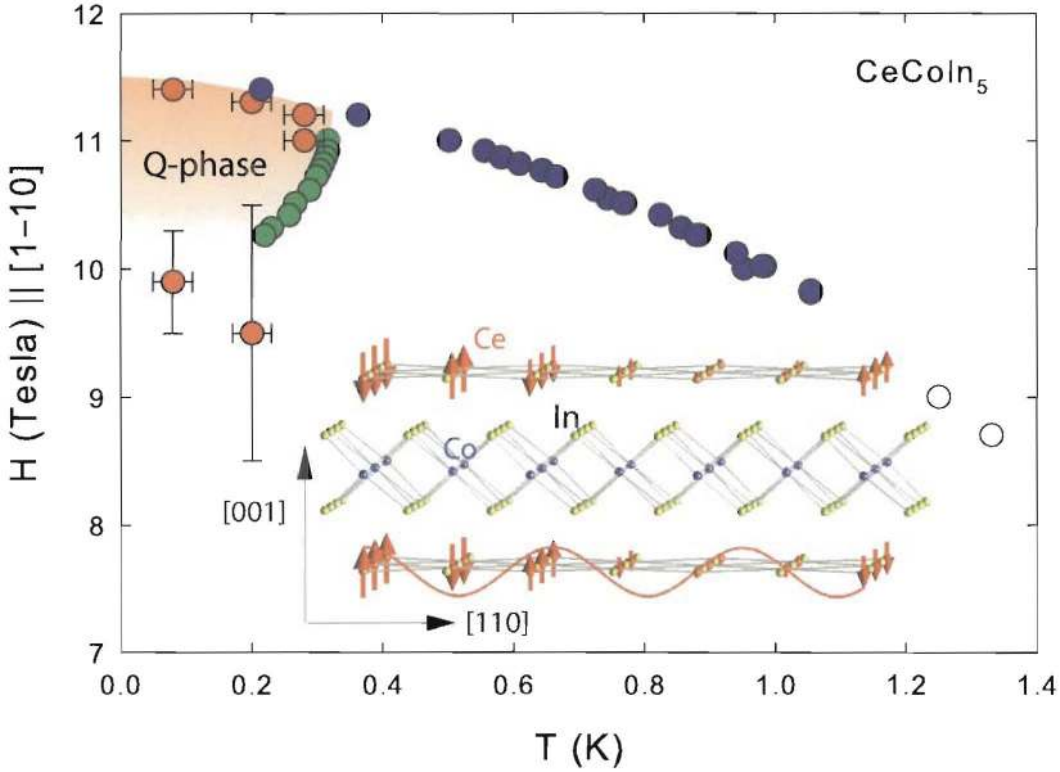

CeCoIn5
Q-phase
H (Tesla) || [1-10]
T (K)
Ce
In
Co
[001]
[110]
12
11
10
9
8
7
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4

Fig. 1

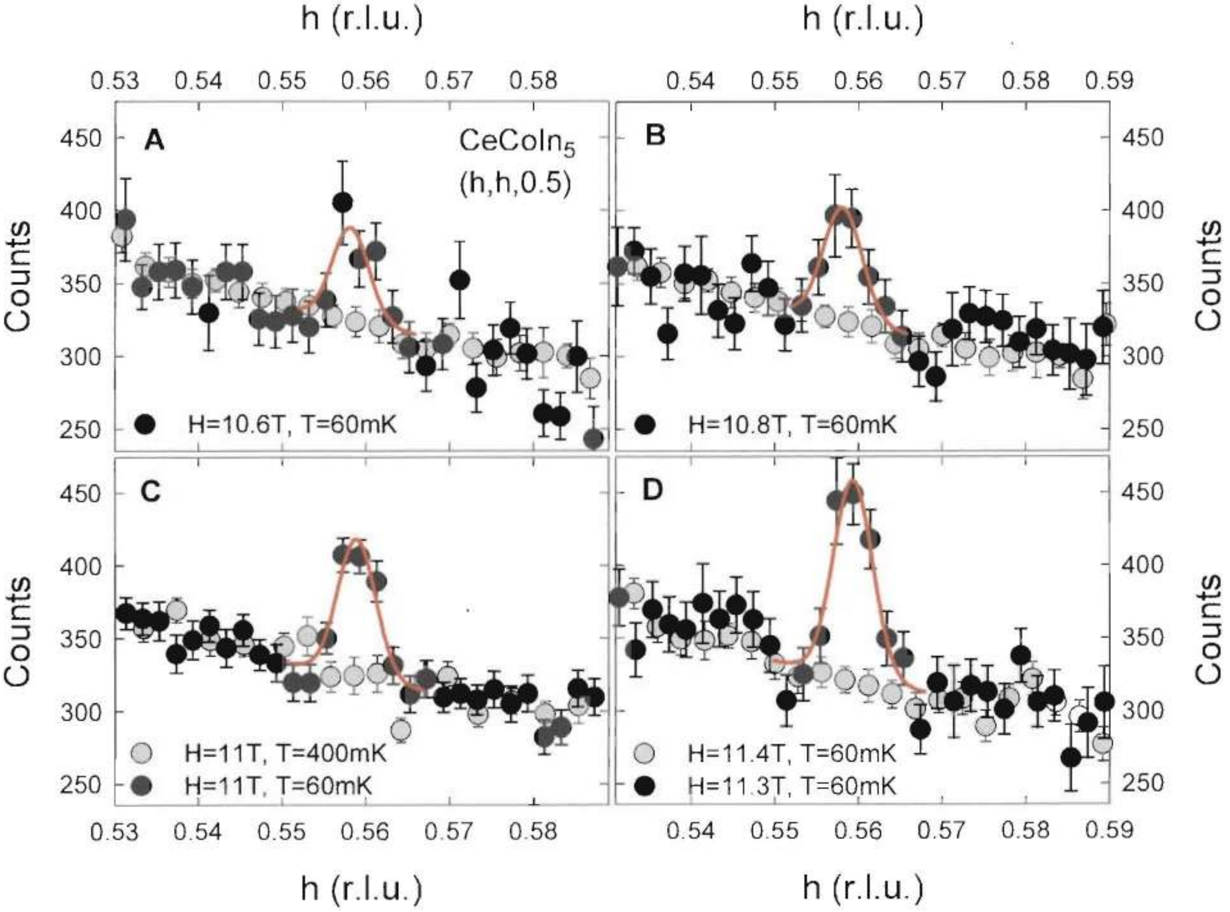
h (r.l.u.)
0.53
0.54
0.55
0.56
0.57
0.58
0.59
Counts
450
400
350
300
250
A
CeCoIn$_5$
(h,h,0.5)
● H=10.6T, T=60mK
B
● H=10.8T, T=60mK
C
● H=11T, T=400mK
● H=11T, T=60mK
D
● H=11.4T, T=60mK
● H=11.3T, T=60mK

Fig. 2

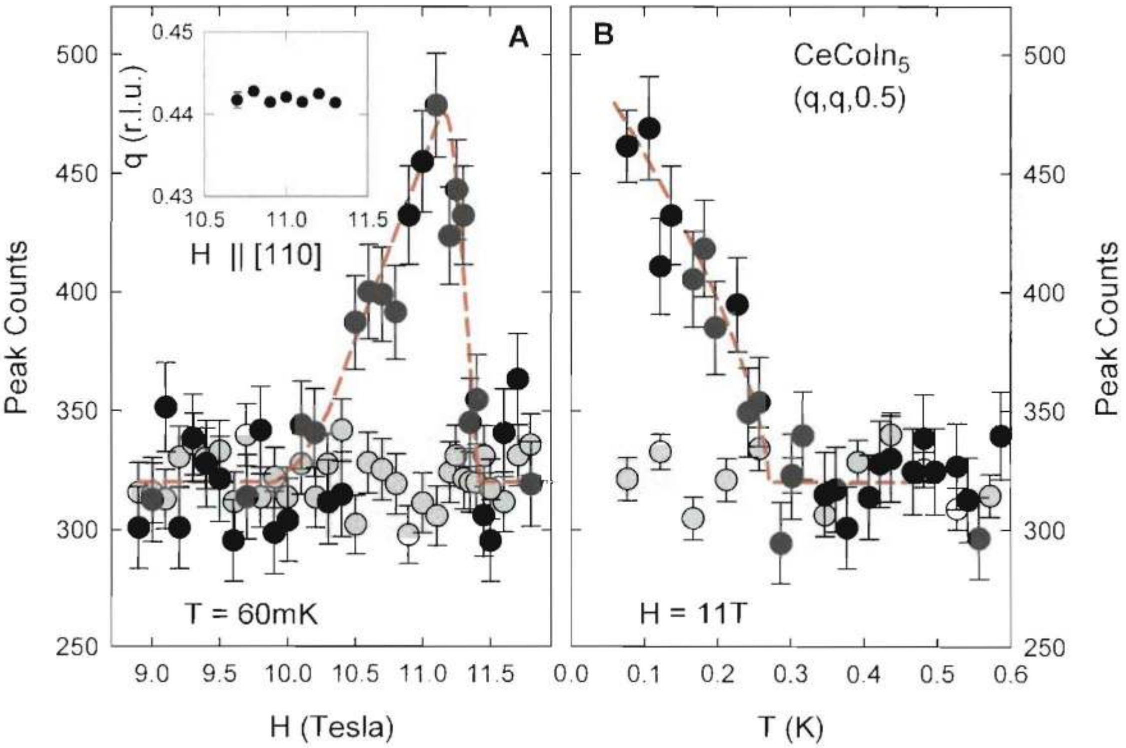
A
B
CeCoIn5
(q,q,0.5)
T = 60mK
H = 11T
Peak Counts
H (Tesla)
T (K)
250
300
350
400
450
500
9.0
9.5
10.0
10.5
11.0
11.5
0.0
0.1
0.2
0.3
0.4
0.5
0.6
q (r.l.u.)
0.43
0.44
0.45
10.5
11.0
11.5
H || [110]

Fig. 3