Ising incommensurate spin resonance of CeCoIn$_5$: a dynamical precursor of the Q-phase.

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(Dated: May 4, 2015)

It is shown by detailed inelastic neutron scattering experiments that the gapped collective magnetic excitation of the unconventional superconductor CeCoIn$_5$, the spin resonance mode, is incommensurate and that the corresponding fluctuations are of Ising nature. The incommensurate peak position of these fluctuations corresponds to the propagation vector of the adjacent field induced static magnetic ordered phase, the so-called Q-phase. Furthermore, the direction of the magnetic moment fluctuations is also the direction of the ordered magnetic moments of the Q-phase. Hence the resonance mode and the Q-phase share the same symmetry and this strongly supports a scenario where the static order is realized by a condensation of the magnetic excitation.

PACS numbers:

Unconventional superconductivity is reported for compounds belonging to various families of materials spanning $d$ and $f$-electron physics: cuprates, iron based materials and heavy fermion systems. Despite this diversity, phenomenological similarities emerge indicating a possible common underlying physics. Firstly, unconventional superconductivity often occurs on the verge of magnetic ordering. Secondly, a universal feedback of unconventional superconductivity manifests on the magnetic excitation spectrum, measured by inelastic neutron scattering, via the appearance of a new well-defined mode in the superconducting phase: the resonance peak. These points motivate a continuous theoretical and experimental effort in the parallel investigations of such systems, in particular in view of the possible common mechanism of spin fluctuation mediated superconductivity. In this context, the study of the relationship between the resonance mode and the adjacent long-range magnetically ordered phase is of first importance and can give insight into the interplay between magnetism and superconductivity. The heavy fermion superconductor CeCoIn$_5$ provides a unique opportunity for such an investigation on an $f$-electron system since both the dynamical resonance mode associated with superconductivity and long range magnetic ordering are reported in this system for zero and respectively finite applied magnetic field.

CeCoIn$_5$ has the highest superconducting transition temperature, $T_c = 2.3$ K, among Ce-based heavy fermion systems. It crystallizes in the tetragonal space group P4/mmm and it is established that the superconducting gap symmetry is of the singlet $d_{x^2-y^2}$ type. In this compound, the spin resonance is observed at the antiferromagnetic wave-vector $Q_{AF}=(0.5, 0.5, 0.5)$ for an energy of 0.6 meV ($\approx 7$ K) that scales approximately with $3k_BT_c$. This intrinsic low energy scale and the strong Pauli-limited superconductivity in CeCoIn$_5$ lead to the observation of a unique behavior, the Zeeman splitting of the resonance under magnetic field. In parallel to these aspects of the spin dynamics, one of the most intriguing properties of CeCoIn$_5$ is the occurrence, above 10.5 T, of magnetic field induced incommensurate magnetic order with $Q_{IC}=(0.45, 0.45, 0.5)$ for a magnetic field applied in the basal plane of the tetragonal structure. Strikingly, this order disappears above the upper critical field $H_{c2}=11.7$ T where superconductivity is suppressed, indicating a strong interplay between the superconductivity and the magnetic ordering. To date the relationship between the spin resonance and the magnetic ordering is tenuous notably due to the mismatch between their characteristic wave-vectors $Q_{AF}$ and $Q_{IC}$. Nonetheless a significant fact is that the extrapolation to zero energy of the lower energy mode of the Zeeman split resonance occurs in the vicinity of the onset of the Q-phase.

In this letter, it is shown by detailed Inelastic Neutron Scattering (INS) experiments that the spin resonance is in fact incommensurate and is peaked at the same wave-vector than the propagation vector of the field induced magnetic order. Furthermore the fluctuations associated with the resonance are found to be polarized along a unique axis, the $c$-axis and this corresponds also to the direction of the ordered magnetic moments in the sine-wave modulation of the Q-phase. The fact that the resonance mode and the Q-phase have the same symmetry indicates that the former is a dynamical precursor of the latter and this strongly supports a scenario where the static order is realized by a condensation of the magnetic excitation. Such a mechanism falls in a broader range of condensed matter physics phenomena, associated with the so-called soft mode behavior, and ranging from lattice dynamical instability in ferroelectrics to Bose-Einstein condensation of magnons in magnetic insulators.

The experiments were performed on the recently upgraded cold neutron three axis spectrometer IN12 at ILL, Grenoble. The initial neutron beam is provided by a
double focusing pyrolitic graphite (PG) monochroma-
tor. Higher order contamination is removed before the
monochromator by a velocity selector. The spectrometer
was setup in W configuration with $\alpha_{1}$-open-open colli-
mations. For the experimental configuration A, a fixed
$k_{f}=1.3$ Å$^{-1}$ is used with $\alpha_{1}=80'$ and a horizontally
focusing PG analyzer is used. For the experimental con-
figuration B, a fixed $k_{f}=1.15$ Å$^{-1}$ is used with $\alpha_{1}=80'$
and the PG analyzer is kept in flat mode (no horizontal
focalization). For the experimental configuration C,
the incident neutron beam spin state is prepared by a
polarizing cavity located 30 m upstream the instrument
and after the velocity selector. Guide fields, that main-
tain the polarization, are installed all along the neutron
path including around the PG monochromator. A Mezei
flipper is placed before the sample table in order to re-
verse the incident polarization. At the sample position,
a Helmholtz coil is used to define the direction of the
polarization. The scattered beam is analyzed by a com-
bination of a Mezei flipper and an horizontally focusing
Heusler analyzer set at fixed $k_{f}=1.3$ Å$^{-1}$, $\alpha_{1}=\text{open}
for this setup. The flipping ratio measured on the (1,1,1)
and (1,1,0) Bragg peaks at $T=1.45$ K in the supercon-
ducting phase for the three polarization channels and the
two flippers varies between 15 and 19. No polarization
correction is applied to the data. The sample is similar
to the one used in our previous studies \cite{10, 14} with an
increased volume. It is composed of an assembly of about
80 single crystals with a total volume of about 250 mm$^{3}$.
The total mosaicity of this assembly is about 1.8 degree.
The sample was put in a He flow cryostat with the [1, -1,
0] axis vertical and the base temperature was 1.45 K.

INS experiments allow to determine the spin dynam-
ics as a function of the wave-vector $\mathbf{Q}$ and the energy $E$
. In the present paper, the wave-vector $\mathbf{Q}$ has its car-
tesian coordinates ($Q_{H}$, $Q_{H}$, $Q_{L}$) expressed in reciprocal
lattice units (r.l.u.). The spin resonance was precisely
mapped out in the region of ($\mathbf{Q}$, $E$) space correspond-
ing to its known maximum intensity with the experimen-
tal configuration A. Focus was given on the ($Q_{H}$, $Q_{H}$,
0.5) direction that spans both $\mathbf{Q}_{\text{AF}}$ and $\mathbf{Q}_{\text{IC}}$. Figure
1 shows a color-coded intensity plot of the INS spectra
collected as a function of $Q_{H}$ and $E$ at 1.45 K. The spin
resonance is clearly incommensurate in the whole mea-
sured energy range (0.4-0.7 meV) and a slight upward
dispersion is observed. A cut along ($Q_{H}$, $Q_{H}$, 0.5) at
a fixed energy of 0.5 meV is shown in Figure 2a with
an increased $\mathbf{Q}$-resolution allowing to better resolve the
incommensurate peaks (Experimental configuration B).
Similar scans performed along the $c$-axis show that the
signal is commensurate in this direction (Figure 2b).
A global fit of the data shown in Figure 2, using Lorentzian
lineshapes, gives the position of the peaks at $\mathbf{Q}_{\text{AF}} \pm (\delta,
\delta, 0)$ with $\delta=0.042 (2)$ r.l.u.. This value of the incommen-
suration determined on the inelastic correlation peaks is
slightly lower than the one ($\delta_{Q}=0.05(1)$) determined on
narrow elastic Bragg peaks for the $\mathbf{Q}$-phase \cite{13}. Setting $\delta = \delta_{Q}$ also provides an acceptable description of
the data shown in Figure 2a due to the broad nature
of these correlation peaks. Therefore, it is considered
that, within the experimental accuracy, the character-
istic wave-vector of the maximum intensity of the spin
fluctuations corresponds to the propagation vector of the
ordered $\mathbf{Q}$-phase. For completeness, it must be noticed
that $\mathbf{Q}_{\text{IC}}$ is also the propagation vector for the mag-
netic ordering in slightly Nd-substituted CeCoIn$_{5}$ with
$T_{N} < T_{c}$ \cite{12}. The obtained correlation length for the
fluctuations at 0.5 meV are $\xi_{a}=20(3)$ Å and $\xi_{c}=12(1)$ Å.
The correlation along the $a$-axis is larger than the one
previously reported \cite{7} \cite{14} due to the separation of two
incommensurate peaks that were previously reported as
a single peak centered at $\mathbf{Q}_{\text{AF}}$. Still the conclusion that
the magnetic correlation length is smaller than the su-
perconducting coherence length of about 40 Å applies.
Considering a local linear approximation for the upward
dispersion leads to a speed of the excitation of about 4.9
meVÅ, which is to be related to the small exchange in-
teractions of typically 0.5 meV characteristics of similar
Ce-based heavy fermion systems \cite{16} \cite{17}.

In order to determine unambiguously the spin anisotropy
associated with the resonance, polarized neu-
tron experiments were undertaken. Because of the loss
of intensity inherent to this setup, the wave-vector res-
olution is relaxed compared to the measurements shown
above so that the splitting into incommensurate peaks is

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Color-coded intensity plot of the INS spectra of CeCoIn$_{5}$ as a function of $Q_{H}$ and $E$ for $Q=(Q_{H}, Q_{H}, 0.5)$ at 1.45 K (Experimental configuration A). The intensity is given for a counting time of 20 minutes. The empty circles indicate positions where data were collected.}
\end{figure}
not resolved and the data are hence collected for \( Q_{AF} \) as in the previous studies. For the polarized INS cross-sections, the canonical right-handed coordinate system is used with \( x \) along the scattering vector \( Q \), \( y \) perpendicular to \( Q \) in the scattering plane and \( z \) perpendicular to the scattering plane. In the polarized neutron cross-section, Spin Flip (SF) scattering refers to scattering process for which the final polarization is antiparallel to the initial one. The measured double differential neutron cross-section for SF scattering and polarization along the axis \( \alpha \), is written \( \sigma^{SF}_{\alpha} \) with:

\[
\sigma^{SF}_{\alpha} \propto B G_{SF} + 1.39 M^a_{Q,\omega} + 0.92 M^c_{Q,\omega} \tag{1}
\]

\[
\sigma^{SF}_{y} \propto B G_{SF} + M^a_{Q,\omega} \tag{2}
\]

\[
\sigma^{SF}_{z} \propto B G_{SF} + 0.39 M^a_{Q,\omega} + 0.92 M^c_{Q,\omega} \tag{3}
\]

where \( B G_{SF} \) is the background for SF channel that includes for convenience the contributions from nuclear spin scattering and \( M^\beta_{Q,\omega} = \frac{1}{2 \pi} \int < M(-Q)^\beta(0)M(Q)^\beta(t) > e^{-i\omega t}dt \) where \( M(Q)^\beta(t) \) is the \( \beta \) component in the sample frame \((\beta=a, c)\) of the Fourier component of the sample magnetization perpendicular to \( Q \) and \( < .. > \) is the quantum statistical expectation value. The conversion between the cartesian \( x, y, z \) coordinates and the crystal axes \([1,0,0], [0,1,0] \) and \([0,0,1] \) is made considering the angle of 23° between the \([1,1,1] \) and the \([1,1,0] \) directions and making the hypothesis of isotropic fluctuation in the plane (equivalence between \([1,0,0], [0,1,0] \) and \([1,1,0] \) directions).

Figure 2 shows an energy spectra measured at 1.45 K at \( Q_{AF} \) for SF scattering using the Mezei flipper located after the sample with the polarization along \( x \), \( y \) and \( z \). The spin resonance excitation is observed for the polarization along the \( x \) and \( z \) axis with the same intensity while the scattering for the polarization along the \( y \) axis is structureless and corresponds to the background.

Fits of the data are performed using an \( \omega \)-Lorentzian as in a previous work [14] and consistently, the energy of the resonance is found to be \( \Omega_{res} = 0.54(1) \) meV and the linewidth \( \Gamma = 0.07(1) \) meV, which is about twice the resolution. An inspection of these data in view of Eq.(1)-(3) indicates that \( M^a_{Q,\omega} \) is zero and \( M^c_{Q,\omega} \) is non vanishing. Hence the fluctuations associated with the resonance peak are polarized along the \( c \)-axis of the tetragonal structure without any in-plane contribution. The \( c \)-axis is also the direction of the amplitude modulated ordered magnetic moments in the magnetic field induced \( Q \)-phase. The exact same conclusions are reached for the data (not shown) collected in the Non Spin Flip channel.

In this paper, it is established by INS that (i) the spin resonance of CeCoIn$_5$ is incommensurate and located at \( Q_{IC} \) (ii) the corresponding fluctuations are polarized solely along the \( c \)-axis. These two characteristic features are shared with the \( Q \)-phase : it is an incommensurate sine-wave modulated structure of propagation vector \( Q_{IC} \) and the magnetic moments are ordered along the \( c \)-axis. The fact that the dynamical mode at \( H=0 \) \( T \) and the field induced static order share to the same symmetry underlines the fact that the resonance is a dynamical precursor of the \( Q \)-phase. This, together with the known softening of the lowest energy mode of the Zeeman split resonance [9][10][14], strongly supports the theoretical scenario where the magnetic ordering is obtained by a field induced condensation of the resonance. Such a scenario was theoretically put forward in a microscopic model by Michal and Mineev [18] but, up to the present study, there was (i) an important mismatch between the characteristic wave-vector of the spin dynam-
Field induced or field-enhanced magnetic ordering out of a superconducting phase is well-known for cuprates [20]. The soft-mode scenario was also considered on phenomenological ground for La$_{1.855}$Sr$_{0.145}$CuO$_4$ where the low energy spin-gap decreases with magnetic field and collapses when antiferromagnetism appears [21]. A common view for cuprates is that the magnetic field reveals the underlying ground state of the normal (non-superconducting) phase in a generic quantum critical phase diagram [22]. This view is in stark contrast with the situation of CeCoIn$_5$ where the cooperative nature of the interaction between magnetism and $d$-wave superconductivity is already obvious from the disappearance of magnetic ordering at $H_{c2}$ and their tight microscopic relation is furthermore highlighted by the pinning of $Q_{IC}$ to the superconducting gap nodal direction [13] and the switching of magneto-superconducting domains [23].

The specificities of a $4f$-electron system like CeCoIn$_5$ opens the way to new aspects absent for the $d$-electron based unconventional superconductors and this can help understanding more generally the nature of the spin resonance and consequently the unconventional superconducting state itself. However, the nature of the resonance peak is not completely settled and a spin-exciton [24], the most common model [25], or a magnon-like excitation [26] are proposed for CeCoIn$_5$. Four main characteristics of the spin resonance of CeCoIn$_5$ are evidenced in the present work, namely the incommensurability, the upward dispersion, the modest exchange and the uniaxial fluctuations. The first three characteristics are shared with the resonance mode of CeCu$_2$Si$_2$ [27], suggesting possible distinctive features of the excitation spectrum of paramagnetic heavy fermion superconductors. These specificities, reminiscent of "heavy-fermion physics", underline the role of the Fermi surface topology and the RKKY nature of the magnetic interactions. While the Fermi surface is composed of multiple quasi-two-dimensional sheets [28], a single band is pointed out to be the relevant one when wave-vectors in the vicinity of $Q_{AF}$ are involved [11]. However to date and despite their increasing accuracy, experimental and theoretical band structure determinations of CeCoIn$_5$ do not reveal the fine details of the Fermi surface topology that define the vector $Q_{IC}$ [29]. Because of this lack of knowledge, it seems premature to draw any conclusion from the observed upward dispersion that is predicted in the magnon-like model [26] while a downward dispersion is predicted for the spin-exciton model [24]. Indeed both models consider a commensurate resonance located at $Q_{AF}$. Our finding of incommensurate resonance therefore asks to revisit these models and their corresponding dispersion. Finally, it must be underlined that the Ising nature of the fluctuations is unique among resonance excitations in unconventional superconductors and arises from the crystal field anisotropy governing such $4f$-electron systems. In contrast, for $d$-electron based unconventional superconductors, where the origin of anisotropy is the spin-orbit coupling, polarized INS data reveal an overall modest splitting in energy of in-plane and out-of-plane components of the fluctuations for Fe-based superconductors [30] or cuprates [31].

The new relationship between the spin resonance excitation and the ordered Q-phase of CeCoIn$_5$ that evidences their common symmetry strongly supports the scenario where the Q-phase is obtained by a spin excitation condensation under magnetic field. This underlines the application of the generic concept, transverse to condensed matter physics, of soft mode for a complex system where magnetism and superconductivity are strongly interconnected.

We acknowledge L.P. Regnault, Y. Sidis and V.P. Mineev for illuminating discussions and K. Mony and B. Vettard for their technical support during sample preparation and neutron scattering experiment.

[1] Y.J. Uemura, Nature Materials 8, 253 (2009).
[2] D.J. Scalapino, Rev. Mod. Phys. 84, 1383 (2012).
[3] Yu, G., Motoyama, E.M. and Greven, Nature Physics 5, 873-875 (2009).
[4] C. Petrovic et al., J. Phys. Condensed Matter 13, L337 (2001).
[5] M.P. Allan et al., Nature Physics 9, 468 (2013).
[6] B.B. Zhou et al., Nature Physics 9, 474 (2013).
[7] C. Stock et al., Phys. Rev. Lett. 100, 087001 (2008).
[8] A. Bianchi et al., Phys. Rev. Lett. 91, 187004 (2003).
[9] C. Stock et al., Phys. Rev. Lett. 109, 167207 (2012).
[10] S. Raymond et al., Phys. Rev. Lett. 109, 237210 (2012).
[11] A. Akbari and P. Thalmeier, Phys. Rev. B 86,134516 (2012).
[12] M. Kenzelmann et al., Science 321, 1652-1654 (2008).
[13] M. Kenzelmann et al., Phys. Rev. Lett. 104, 127001 (2010).
[14] J. Panarin et al., J. Phys. Soc. Japan, 78, 113706 (2009).
[15] S. Raymond et al., J. Phys. Soc. Japan 83, 013707 (2014).
[16] W. Knafo et al., J. Phys. : Condensed Matter 15, 3741 (2003).
[17] P. Das et al., Phys. Rev. Lett. 113, 264603 (2014).
[18] V.P. Michal and V.P. Mineev, Phys. Rev. B 84, 052508 (2011).
[19] The first unpolarized INS study of Stock et al. (Ref. [5]) suggests that the spin fluctuations are polarized along the c-axis from the dependence of the signal in several Brillouin zone. However the observed signal decreases faster than the calculation which casted some doubt on the assumption (See Fig.2 of Ref.[5]).
[20] B. Lake et al., Nature 415 299 (2002).
[21] J. Chang et al., Phys. Rev. Lett. 102, 177006 (2009).
[22] E. Demler et al., Phys. Rev. Lett. 87 067202 (2001).
[23] S. Gerber et al., Nature Physics 10, 126 (2014).
[24] E. Eremin et al., Phys. Rev. Lett. 101, 187001 (2008).
[25] M. Eschrig, Adv. Phys. 55, 47 (2006) and references therein.
[26] A.V. Chubukov and L.P. Gor’kov, Phys. Rev. Lett. 101, 147004 (2008).
[27] O. Stockert et al., Nature Physics 7 (2011) 119.
[28] R. Settai et al., J. Phys. Condens. Matter 13, L627 (2001).
[29] T. Nomoto and H. Ikeda, Phys. Rev. B 90, 125147 (2014).
[30] D.S. Inosov, Comptes Rendus Physique preprint : arXiv:1502.06570.
[31] N.S. Headings et al., Phys. Rev. B 84, 104513 (2011).