Effects of impurities in CeCoIn$_5$ using inelastic neutron scattering

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Abstract. The influence of Nd magnetic impurities on the spin dynamics of CeCoIn$_5$ was studied by inelastic neutron scattering. In the Nd-substituted compound Ce$_{0.95}$Nd$_{0.05}$CoIn$_5$, the spin resonance peak (observed at $\Omega_{\text{res}} = 0.55$ meV in the pure system) is shifted to lower energies but the ratio $\Omega_{\text{res}}/k_B T_c$ remains almost unchanged. These observations are compared with non-magnetic La-substitution.

1. Introduction

Superconductivity is a macroscopic quantum state resulting from the condensation of electrons in Cooper pairs. In the case of conventional superconductivity the pairing mechanism is the weak electron-phonon interaction. However in strongly correlated systems exhibiting a superconducting (SC) state the pairing mechanism is thought to be of a different nature. Examples of such unconventional superconductors are high-temperature superconducting cuprates (HTSC), heavy-fermion compounds (HF) and the iron-based superconductors. In these compounds, the origin of the pairing is strongly suspected to be the magnetic interaction, and a seminal study of the magnetic excitation spectra by inelastic neutron scattering (INS) in the cuprate YBa$_2$Cu$_3$O$_{6+x}$ showed the appearance of a sharp excitation called a magnetic spin resonance [1] in the SC state. This was later generalized to other cuprates. Such a feedback of superconductivity on the magnetic excitation spectra was backed up by theories of a pairing mechanism of magnetic origin. The recent discovery of similar excitations in HF superconductors UPd$_2$Al$_3$[2] CeCoIn$_5$[3] and CeCu$_2$Si$_2$[4], as well as in the iron superconductors [5] suggests that the magnetic resonance could be a universal feature of unconventional superconductors.

Among HF superconductors the compound CeCoIn$_5$ has the highest critical temperature, $T_c = 2.3$ K [6]. It crystallizes in the tetragonal space group $P4/mmm$ and consists of alternating CeIn$_3$ and CoIn$_2$ layers. A quasi-2D nature is supported by de Haas van Alphen measurements, which established a Fermi surface composed of nearly cylindrical sheets [7]. As concerns the low-energy magnetic excitations measured by INS, a quasi-elastic signal is measured above $T_c$ with a linewidth of 0.3 meV. Below $T_c$, the spectral shape changes from a quasi-elastic to a sharp inelastic peak, which appears at an energy of $\Omega_{\text{res}} \approx 0.55$ meV ($\approx 2.7k_B T_c$) at the antiferromagnetic position $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ [3]. The introduction of impurities is a useful probe to investigate the microscopic nature of the SC state [8]. For a $d$-wave superconductor like CeCoIn$_5$, impurities are expected to have a Cooper pair breaking effect [9]. In CeCoIn$_5$ the substitution of other lanthanid on the Ce site make it
possible to probe the effect of non-magnetic (La)\([10][11]\) and magnetic impurities (Nd)\([12]\) on the spin resonance. The effect of these impurities on the SC state differs from those observed in conventional superconductors for which non-magnetic impurities (at low concentration) do not affect the superconductivity. Indeed La \((4f^0)\) reduces the critical temperature \(T_c\) by \((-0.056T_c)/(1 \% \text{ of La substitution})\)\([10]\), the Nd\((4f^0)\) reduces \(T_c\) by \((-0.042T_c)/(1\% \text{ of Nd substitution})\). Moreover Nd induces a magnetic order within the SC state at very low temperature. The ordering temperature \(T_m\) increases with Nd substitution \([12]\).

2. Experimental details
In this article, we report INS experiments performed on single crystal samples of Ce\(_{0.95}\)Nd\(_{0.05}\)CoIn\(_5\) and compared the results with those obtained in Ref.\([13]\) for Ce\(_{0.98}\)La\(_{0.02}\)CoIn\(_5\). We find that the ratio between the resonance energy \(\Omega_{\text{res}}\) and \(T_c\) remains almost constant for Nd impurities as found previously from La impurities but the excitation broadening is less pronounced with Nd substitution than with La substitution.

Single-crystal samples of Ce\(_{0.95}\)Nd\(_{0.05}\)CoIn\(_5\) were grown by the self-flux method \([14]\) and characterized by specific heat measurements performed using a commercial Quantum Design Physical Properties Measurement System (PPMS) down to \(T = 400\) mK. From these measurements we deduce the \(T_c\) which is equal to 1.77 K. For this composition, the magnetic ordering temperature \(T_m\) is around 0.8 K.

The INS experiment was performed on the cold neutron triple-axis spectrometer 4F1 at the Laboratoire Léon Brillouin, Saclay. In this experiment, the incident beam was provided by a double pyrolytic graphite (PG) monochromator. A liquid-nitrogen-cooled Be filter was placed before the sample in order to avoid any higher order contaminations. Measurements were performed with a fixed final wavevector \(k_f = 1.3\) Å\(^{-1}\). The collimations were 60'-open-open. The energy resolution determined from the full width at half-maximum (FWHM) of the incoherent signal was 0.16 meV. The sample consisted of an assembly of about 20 single crystals of Ce\(_{0.95}\)Nd\(_{0.05}\)CoIn\(_5\) co-aligned and glued with Fomblin oil on two thin aluminum plates. The mosaic spread of the assembly, as derived from a rocking curve through the (1,1,1) Bragg reflection, was around 1 degree. The sample was put in a \(^3\)He-\(^4\)He dilution insert with \([1 1 0]\) and \([0 0 1]\) axes defining the scattering plane.

In a triple-axis experiment performed at constant \(k_f\), the background corrected neutron intensity is proportional to the scattering function \(S(Q, E)\), which is itself proportional to the imaginary part of the dynamic susceptibility \(\chi''(Q, E)\),

\[
S(Q, E) = n(E, T) \chi''(Q, E)
\]

\(\chi''\) was analysed using an inelastic Lorentzian spectral function where \(n(E, T) = 1/(1 - e^{-E/k_BT})\) is the detailed balance factor,

\[
\chi''(Q, E) = \frac{1}{2} \left( \frac{\chi_Q \Gamma_Q E}{(E - \Omega_{\text{res}})^2 + \Gamma_Q^2} + \frac{\chi_Q \Gamma_Q E}{(E + \Omega_{\text{res}})^2 + \Gamma_Q^2} \right)
\]

Here \(\Gamma_Q\) is the relaxation rate, \(\Omega_{\text{res}}\) the resonance energy and \(\chi_Q\) the static susceptibility at the wave-vector \(Q\).

3. Results and Discussion
The magnetic excitation spectrum measured at \(Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})\) is shown in Fig.1 in the upper panel. The dashed line corresponds to the background signal obtained by performing an energy scan at a different \(Q\) vector, \(Q = (\frac{1}{2}, \frac{1}{2}, 0.2)\). Without substitution, the resonance peak was reported \([15]\) to occur at 0.55 meV, with a relaxation rate of 0.07 meV. In the 5% Nd-substituted
system the resonance peak shifts to $\Omega_{res} = 0.41$ meV and undergoes a weak broadening, $\Gamma_Q = 0.10 \pm 0.012$ meV. In consequence the ratio $\Omega_{res}/k_B T_c$ is about 2.7 as in the pure compound. For comparison, the lower panel of Fig.1 shows the spectrum obtained on a 2% La-substituted compound reported in [13] with $T_c = 1.9$ K close to the $T_c$ of Ce$_{0.95}$Nd$_{0.05}$CoIn$_5$ ($T_c = 1.8$ K). For that La-concentration, $\Omega_{res}$ is equal to 0.45 meV but the linewidth is substantially increased, with $\Gamma_Q = 0.15 \pm 0.01$ meV. For a 3.5% La-substituted compound (not shown here) with $T_c = 1.7$ K, $\Omega_{res}$ is equal to 0.35 meV and the linewidth is also $\Gamma_Q = 0.15 \pm 0.02$ meV.

A constant-energy scan was performed along the $c$-axis around $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ in order to measure the correlation length of the resonance upon Nd-substitution. Fig.2 shows the spectrum measured at $E = 0.45$ meV. The scan is analyzed using a Gaussian lineshape. The background is determined by measurements at high temperature where the magnetic spectrum is no longer peaked in $Q$-space. The signal still show a maximum at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, as for the pure compound. The correlation length obtained from the inverse of the Gaussian half-width at half maximum is $\xi_c = 6.5 \pm 0.1$ Å exactly as in the pure compound [15]. In the 2% La-substituted sample $\xi_c$ shows a small decrease with a value of $\xi_c = 5.1 \pm 0.1$ Å. A constant energy scan performed along the $a$-axis cannot be performed for the 5%-substituted compound due to geometrical constraints of the spectrometer limiting the range in $Q$-space.

The comparison between Nd and La substitutions can be summarized as follow. The evolution
of the linewidth is different in the two cases. Indeed whereas the La-substitution induces a substantial broadening, Nd-substitution weakly increases the width of the excitation. Such effect means that non-magnetic impurities have a stronger effect on the life-time of the spin resonance. However both substituents decrease the resonance energy while conserving the ratio Ω_{res}/k_BT_c. This decrease could be explained by an impurity effect on the SC gap [9] leading to a conservation of this ratio as it was proposed in Ref.[13] for La-substitution. To our knowledge the only INS experiment showing the effect of magnetic and non-magnetic impurities on the spin resonance [16] compared the effects of non-magnetic Zn and magnetic Ni substitution in YBa_2Cu_3O_7. A detailed comparison between that experiment and La-substitution was presented in Ref.[13]. There we could complete this parallel for magnetic impurities in CeCoIn_5. As in the cuprate, the magnetic substitution decreases T_c less than its non-magnetic equivalent. Moreover, both impurities have no effect on the ratio Ω_{res}/k_BT_c and no influence on the correlation lengths. Whereas there is stark contrast between La and Zn substitution [13], the effect of magnetic impurities in CeCoIn_5 and YBa_2Cu_3O_7 seem very similar reinforcing the idea that magnetic impurities do not disrupt spin correlations over large scales [16].

4. Conclusion
Our INS experiments showed that the substitution of magnetic Nd impurity in CeCoIn_5 has one pmaineffect on the spin resonance: a decrease the resonance energy which is interpreted by a SC gap diminution. Contrary to the non-magnetic case, the resonance shows a very weak broadening indicating that the spin resonance is weakly affected by magnetic impurities. The next step of this work will be to performed INS experiments at low temperatures to observe the evolution of the spin resonance in the magnetically ordered phase and to extend the measurements to other Nd concentration to confirm the present results.

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