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## Ce<sub>3</sub>Pt<sub>23</sub>Si<sub>11</sub> ferromagnet: unusual magnetic behavior brought to light by neutron studies

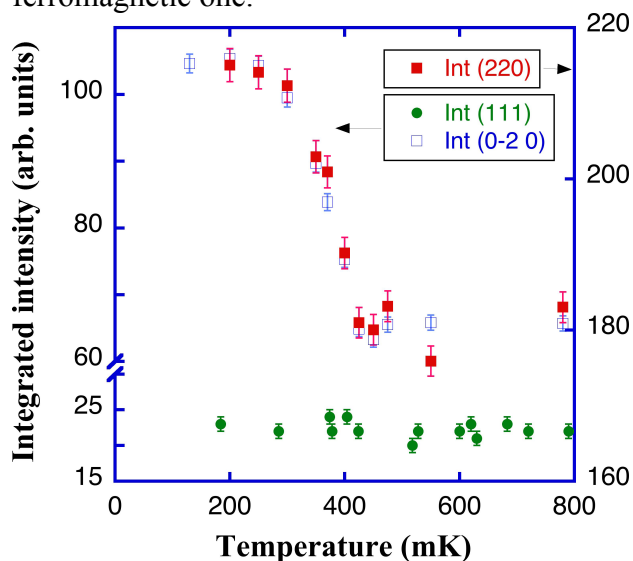
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S. Rols and B. Ouladdiaf (ILL)

Instruments : Four-circle diffractometer D10  
Time-of-flight spectrometer IN4

### Introduction :

The ternary system R-Pt-Si (R = rare earth element) triggers a lot of interest, due to peculiar physical behaviors of some compounds: unconventional superconductivity, heavy-fermion properties, coexistence of superconductivity and magnetic ordering..., such as CePt<sub>3</sub>Si, reported as the first non-centro symmetric heavy fermion superconductor. Actually, several studies let suppose a sample dependence of the superconductivity and for instance, in Ce deficient samples, the coexistence of a new phase, Ce<sub>3</sub>Pt<sub>23</sub>Si<sub>11</sub>, was suspected to interfere with superconducting properties. We have undertaken an extended investigation on this new phase.

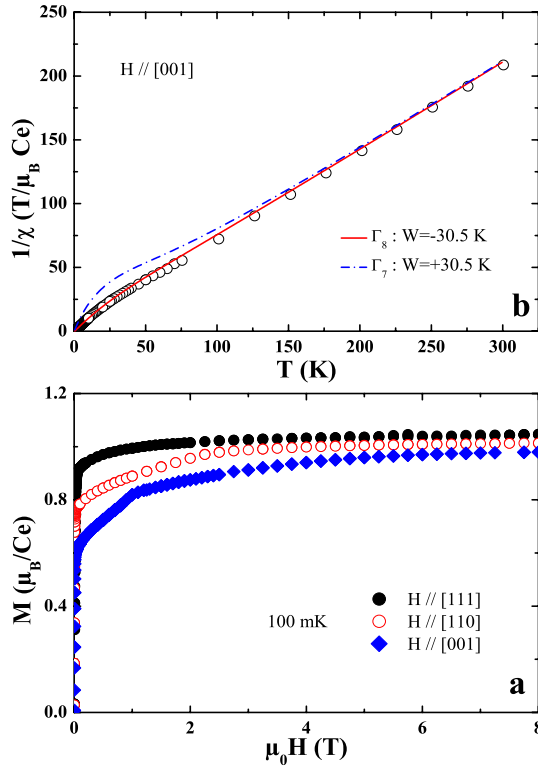
Ce<sub>3</sub>Pt<sub>23</sub>Si<sub>11</sub> crystallizes in a face-centered cubic structure (Fm-3m space group). Recently, we have synthesized very high quality single crystals. Magnetic measurements have shown that Ce<sub>3</sub>Pt<sub>23</sub>Si<sub>11</sub> orders ferromagnetically at  $T_c = 0.44$  K with a spontaneous magnetic moment far smaller than the theoretical value of  $2.14 \mu_B/\text{Ce}$  expected for Ce<sup>3+</sup> ion [1]. Such a reduction may arise from crystal electric field (CEF) effect and / or from magnetic arrangements more complex than the collinear ferromagnetic one.



**Figure 1:** Thermal variation of the integrated intensity of the (220), (0-20) and (111) reflections (error bars are represented for each data point).

Neutron diffraction experiments performed on a single crystal using the unique four-circle dilution cryostat available on the D10 diffractometer, confirmed the transition at  $T_c$ . Below  $T_c$ , as illustrated by the temperature dependence of the (220), (0-20) and (111) reflections (figure 1), the magnetic contribution is only observed on top of nuclear reflections with hkl all even. The non-observation of a magnetic contribution for odd reflections indices is a strong constraint. This leads to ferromagnetic couplings between pairs of Ce ions within the 6 Ce ions in the unit cell. Group theory analysis shows that the only non-zero basis vectors, consistent with a ferromagnetic arrangement on the Ce sites are those of the  $\Gamma_{10}$  irreducible representation. These vectors describe a ferromagnetic arrangement along [100], [010] and [001] axis respectively, while a combination of the three basis vectors leads to a ferromagnetic arrangement along the [111] direction. This is in very good agreement with the magnetization process, shown in figure 2a. The refined value of the magnetic moment,  $1.2(2)\mu_B$  per Ce ion, is close to that deduced from the magnetization measurements (figure 2a). Both determinations strongly indicate a doublet CEF ground state.

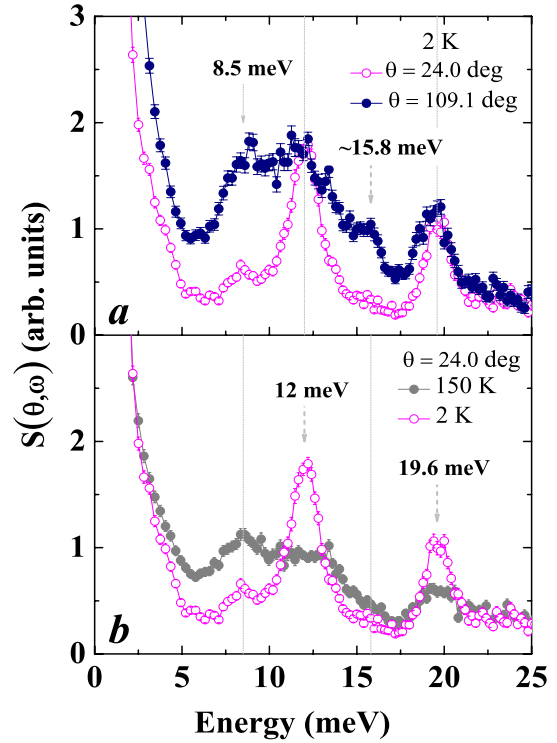
The CEF excitations were studied by inelastic



**Figure 2:** a) Magnetization curves along the three principle axes of the cube in the ferromagnetic phase at 100mK. b) Thermal variation of the inverse of the magnetic susceptibility ( $\mu_0 H // [001]$ ). The lines represent the calculated curves using the values of the CEF parameter  $W$  reported in the figure.

neutron scattering on the IN4 time-of-flight spectrometer. For  $\text{Ce}^{3+}$  ions in cubic symmetry, only fourth degree terms are effective in the CEF Hamiltonian and thus only one parameter,  $W = \pm E_{\text{CEF}}/6$  has to be considered [2]. A positive (negative)  $W$  selects a doublet (quadruplet) as CEF ground state. Unexpectedly, two magnetic excitations are observed at  $E_1=12 \text{ meV}$  and  $E_2=19.6 \text{ meV}$  on the spectra (figure 3), while neutron diffraction experiments shows that the structure remains cubic down to 800 mK. Observation of two CEF excitations in a cubic Ce compound was earlier reported in  $\text{CeAl}_2$ . It was interpreted in terms of an unusual magneto-elastic coupling that gives rise to a bounding state between the excited CEF quadruplet level and low-lying vibronic states [3-5].

Calculations of the magnetic susceptibility within the CEF formalism show that experimental data are in better agreement with



**Figure 3:** a) Inelastic intensities for two scattering angles at 2 K. b) Inelastic intensities for  $T = 2 \text{ K}$  and 150 K at low angle detector averaging.

a quadruplet CEF ground state (figure 2b). In this case an interpretation similar to that of  $\text{CeAl}_2$  should not apply, as the excited doublet cannot be split. The discrepancy between the different experimental observations in  $\text{Ce}_3\text{Pt}_{23}\text{Si}_{11}$  together with an apparent crystal symmetry that remains cubic can be tentatively explained by the splitting of the quadruplet ground state via a dynamical Jahn-Teller effect [6]. This hypothesis will need further exploring.

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