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Metamagnetic transition in the Kondo lattice compound $CePt_2Si_2$

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Detailed magnetization measurements have been performed on a single crystal of the tetragonal nonmagnetic Kondo lattice compound $CePt_2Si_2$. A careful analysis of the data reveals an anomalous behavior below 4 K: namely, a strong decrease of the initial susceptibility associated with the existence of a metamagnetic transition at about 30 kOe inside the basal plane.

Anomalous properties have been recently reported on a new Kondo lattice compound, $CePt_2Si_2$.[1,2] Susceptibility measurements, performed on a polycrystalline material, have shown (i) a normal Curie-Weiss behavior at high temperature, although the paramagnetic Curie temperature is strongly negative ($\theta_p = -86$ K), (ii) a broad maximum around 60 K, and (iii) a constant value below 20 K. Resistivity presents a logarithmic decrease at high temperature, a large maximum at 76 K, and a steep decrease below this temperature, following roughly a T^2 law below 15 K. The specific heat has been interpreted as indicating coherence effects below 2 K, temperature where C/T reaches a maximum of 120 $mJ/mol\,K^2$. In the same temperature range, the resistivity has been found to deviate from the T^2 law.[3]

All these results indicate the existence of three regimes in $CePt_2Si_2$, depending on the temperature: (i) above 150 K, an isolated Kondo impurities behavior with a cerium ion in a state close to a 3^+ one, (ii) an intermediate regime (4–150 K) where the low-temperature nonmagnetic Kondo state takes place, and (iii) finally, below 4 K, the onset of a new regime which has been associated with a coherent Kondo state. Preliminary results of resistivity and magnetic measurements performed on a single crystal confirmed this latter change of regime at low temperature and emphasized the strongly anisotropic character of the magnetic properties.[4] Here, we report a detailed analysis of the magnetization processes, in particular below 4 K, along the main symmetry directions of the tetragonal unit cell.

$CePt_2Si_2$ crystallizes in the $CaBe_2Ge_2$-type tetragonal structure (space group $P4/nmm$) with $a = 4.25$ Å and $c = 9.79$ Å.[5] It appears to form congruently with a high melting temperature.[6,7] A single crystal was grown by the Czochralski method from a stoichiometric mixture of pure constituents (99.99% purity cerium). Magnetic measurements were performed by the extraction method in a superconducting coil ($H < 80$ kOe) in the temperature range 1.5–300 K, the magnetic field being applied along the [100] (**a** axis), [110] and [001] (**c** axis) crystallographic directions. Measurements have been extended up to 180 kOe at the Service National des Champs Intenses in Grenoble.

In the whole investigated range, the magnetization remains very weak, with a large anisotropy in favor of the basal plane, the anisotropy being very weak inside this latter (see Fig. 1). This is particularly obvious on the susceptibility curves (see Fig. 2). At high temperature, a Curie-Weiss law is observed, with a slope corresponding to an effective paramagnetic moment $\mu_p = 2.65 \pm 0.1 \mu_B$ and $2.55 \pm 0.1 \mu_B$ along and perpendicular to the **c** axis,

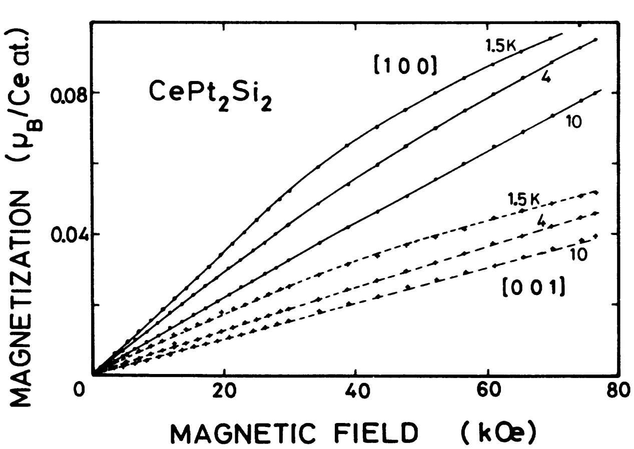

FIG. 1. Total magnetization curves of $CePt_2Si_2$ at several temperatures for [001] and [100] directions. Lines are only guides for eyes.

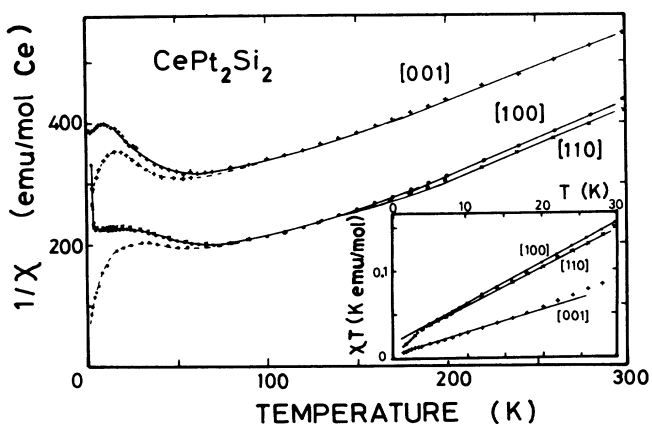

FIG. 2. Magnetic susceptibility of $CePt_2Si_2$ along the three main symmetry directions. Hatched line: raw data; continuous lines: after correction for impurities (see text). Inset: χT vs T curves allowing the separation between intrinsic and impurities effects.

respectively, values very close to that of the Ce^{3+} ion. As for a polycrystalline sample, the paramagnetic Curie temperature is strongly negative, reaching $\theta_p = -175 \pm 10$ K and -50 ± 10 K along the **c** and **a** axes, respectively. This result is in agreement with previous measurements on a single crystal.[7] Assuming that such a splitting between both directions is only due to crystal-field effects, the second-order crystal-field parameter $B_2^0 = +13$ K can be then deduced,[8] leading to a $| \pm \frac{1}{2} \rangle$ crystal-field ground state if this parameter is preponderant.

When decreasing the temperature, the susceptibility exhibits a maximum at 55 and 65 K in the **c** and **a** directions, respectively. The position of these maxima correlates with those observed on the resistivity curves, although the shift between both maxima is larger in this latter case ($T_{max} = 55$ and 85 K, respectively; see Ref. 4). At lower temperature, a noticeable increase of the susceptibility is observed, superimposed to the intrinsic behavior which is expected to saturate. This may be due to a small amount of magnetic impurities, such as "normal" Ce^{3+} ions for example, situated on or near grain boundaries or other defects[9,10] (see below).

In order to properly take into account this impurity contribution, we have performed a careful analysis of the low-field magnetization curves. First, we have assumed a constant intrinsic susceptibility $\chi(0)$ for $CePt_2Si_2$ at low temperature, and a Curie contribution $\alpha C/T$ for the impurities, α being their molar concentration and C the Curie constant of the $J = \frac{5}{2}$ Ce^{3+} ion. A linear behavior of the χT vs T curves is then expected and is effectively observed between 1.5 and 20 K along the **c** axis, and between 4 and 30 K in the basal plane (see Fig. 2). That gives an impurity concentration which is different parallel and perpendicular to the **c** axis ($\alpha = 0.6\%$ and 1.8%, respectively), showing that the impurities seem to be also subjected to crystal-field anisotropy. On the other hand, the slope of these curves provides for $CePt_2Si_2$ the intrinsic susceptibilities $\chi_c(0) = (2.5 \pm 0.1) \times 10^{-3}$ emu/mole and $\chi_a(0) = (4.4 \pm 0.1) \times 10^{-3}$ emu/mole. Note that a small deviation from this linear law is already perceptible below 4 K, but only within the basal plane (see below).

The quantitative determination of these magnetic impurities effects through an elaborate analysis of the whole M_{tot} vs H curves strongly corroborates the above assumption. Indeed, if the field dependence of the intrinsic magnetization is expected to be linear because of the weakness of $\chi(0)$, the magnetic impurities contribution M_{imp} should saturate in high field and at low temperature by following a Brillouin-type law which should scale with H/T:

$$M_{tot} = \chi(0)H + M_{imp}(H/T) \ .$$

Figure 3 shows the impurity contribution obtained by subtracting $\chi(0)H$ from the total magnetization M_{tot} for both [110] and [001] directions. The expected behavior is effectively observed, i.e., a scaling in H/T occurs on M_{imp}, in the same temperature range as the linear law for the χT vs T curves. A consistent description of this impurity contribution, i.e., accounting for its anisotropy by considering a single concentration α for both directions requires to take into account crystal-field effects. Indeed, like the Kondo cerium ions, the Ce^{3+} impurities should be subjected to a crystal field arising from their environment, which splits the sixfold multiplet $J = \frac{5}{2}$ into three doublets. From the available information, it is not possible to unambiguously determine the three crystalline electric field (CEF) parameters involved if the impurities are considered as being in sites with tetragonal symmetry.[8] However, a set of values such as $B_2^0 = 30.7$ K, $B_4^0 = 0.93$ K, and $B_4^4 = 19.5$ K leads to an excellent agreement between calculated and observed variations of M_{imp} vs H/T as well as χ_{imp} vs T (see Fig. 3). An impurity concentration $\alpha = 5.3\%$ is thus obtained. A remarkable feature of these curves is that the anisotropy of Ce^{3+} impurities favors the basal plane as for the Kondo Ce^{3+} ions of bulk $CePt_2Si_2$.

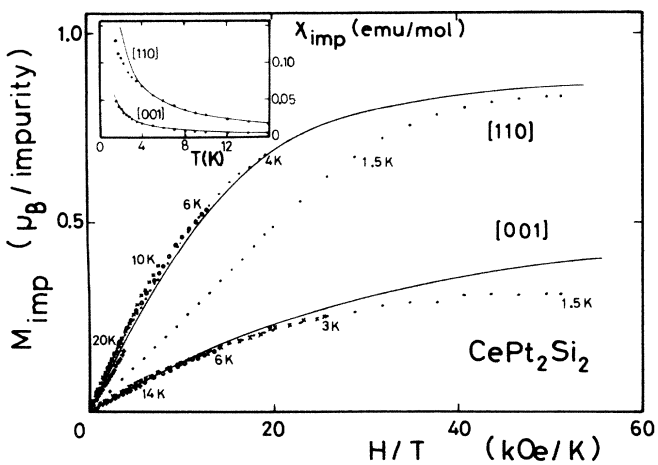

FIG. 3. Impurities contribution to the magnetization vs H/T in $CePt_2Si_2$, after subtracting a linear intrinsic part. Inset: temperature variation of the susceptibility of impurities, assuming an intrinsic susceptibility constant in the range 1.5–15 K. Continuous lines are calculated in a simple crystal-field model as described in the text.

At this stage, as shown in Fig. 3, a net deviation from the Brillouin-type law may be observed below 4 K in the basal plane, and the question is whether this behavior is intrinsic or must be attributed to the impurities themselves. In this latter case, that could be explained by the occurrence of an antiferromagnetic or spin-glass ordering. However, the shape of the impurities susceptibility, within the hypothesis of an intrinsic susceptibility constant down to 1.5 K, exhibits only a slight deviation from a $1/T$ Curie behavior below 4 K along the [110] direction (see inset of Fig. 3). This weak anomaly then cannot be attributed to the onset of such a magnetic ordering for the impurities.

Therefore, the low-temperature deviation in the basal plane is actually an intrinsic property of $CePt_2Si_2$. The Brillouin-type impurity contribution then has been subtracted from the total magnetization at any temperature in order to obtain the actual intrinsic behavior of $CePt_2Si_2$. As shown in Fig. 4, the magnetization processes along the **c** direction present no noticeable change between 1.5 and 10 K. On the contrary, in the basal plane a metamagnetic transition is evidenced below 4 K in the magnetization processes. Especially at 1.5 K, this transition is the most pronounced and occurs for a critical magnetic field of about 30 kOe. As a consequence, a dramatic

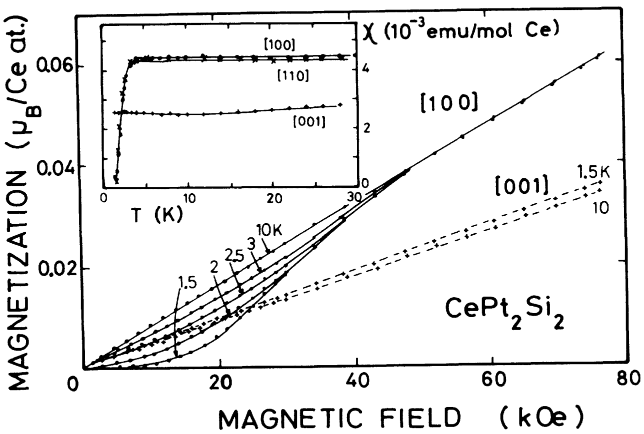

FIG. 4. Low-temperature behavior of intrinsic magnetization curves of $CePt_2Si_2$ along [100] and [001] directions. Inset: temperature variation of the corresponding low-field susceptibility. Lines are only guides for eyes.

collapse of the intrinsic initial susceptibility occurs below 4 K (see inset of Fig. 4). Note that this metamagnetic transition is already perceptible at 1.5 K on the raw magnetization curves (see Fig. 1), and that this low-temperature behavior has been found nearly insensitive to annealing the samples at 1150 °C for three days.

The present results, i.e., the breakdown of the susceptibility and the occurrence of a metamagnetic transition within the basal plane, constitute new evidences of the low-temperature regime previously observed on specific heat and resistivity of $CePt_2Si_2$, where the appearance of a coherent Kondo state was invoked.[2,3] Within this hypothesis, the present phenomenon would be the magnetic signature of the coherent Kondo state. In this case, as it has been theoretically predicted,[11] the critical field would be associated with the destruction of the coherent state by restoring the Kondo lattice state. Another possible explanation could be the existence of a weak antiferromagnetic ordering below 4 K in $CePt_2Si_2$, with a magnetic moment of a few hundredths of a Bohr magneton. This situation is similar to that of the heavy fermion $CeAl_3$, a compound in which the anomalous low-temperature behavior was first interpreted as the entrance in a coherent state[12] and more recently as the manifestation of a possible magnetic ordering.[13] It is worth noting that in $CeAl_3$ the existence of a critical field was evidenced by magnetoresistance and thermopower experiments.[12]

Few detailed magnetization studies are available at the present time on Kondo lattice or heavy-fermion compounds. Generally, the magnetization is small and varies linearly as a function of the magnetic field, leading to a magnetic susceptibility which tends to saturate at low temperature.[14,15] Metamagnetic transitions have been observed in $CeRu_2Si_2$ (Refs. 16 and 17) and UPt_3,[18] but with characteristics different from those in $CePt_2Si_2$: (i) above the transition, the magnetic moment reaches a magnitude much larger than in $CePt_2Si_2$ and closer to the saturated value of the Ce^{3+} or U^{3+} ion, and (ii) the magnitude of the critical field is larger in $CeRu_2Si_2$ and UPt_3.

The metamagnetic behavior of $CePt_2Si_2$ in the basal plane below 4 K may be therefore associated with the disappearance of a low-temperature state which is not entirely understood at this time. Choosing between both possible interpretation—coherent state or antiferromagnetism—is not easy, due to the weak magnitude of the experimental manifestations. In the latter case, the occurrence of a magnetic ordering on a nonmagnetic Kondo state remains to be explained. Other anisotropic features may be expected from different ways of investigation such as magnetoresistance or Hall effect, and a development is underway in that direction.

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