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Pressure effects on the antiferromagnetic order of the hydride CeRuSiH

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Abstract. The non-magnetic heavy fermion CeRuSi upon hydrogen absorption develops an antiferromagnetic behaviour, mainly due to the pronounced expansion of the unit cell. The effects of pressure on the $\chi(T)$ and $M(H)$ curves of the hydride CeRuSiH up to 10.3 kbar are presented. Both, the Néel temperatures and the critical fields of the metamagnetic transitions, increase with pressure, whereas the net magnetization decreases. This evolution could be understood within the framework of the Doniach diagram.

1. Introduction

The insertion of hydrogen provides an interesting means to modify drastically the magnetic properties of many Ce alloys [1]. In some cases, as in the antiferromagnetic CeCoSi, hydrogenation leads to the loss of magnetic order [2] whereas in others, as in CeNiSn, a non-magnetic system transforms into a magnetic one [1, 3]. These changes are mainly due to the competition of two effects. The first one is the modification of electronic structure due to the presence of $1s(H)$ electrons, which results in a strong Ce-H bonding. The second one is due to the change in the unit cell volume, which could be considered as a negative pressure.

Within this framework, hydrogenation of the ternary silicide CeRuSi, which crystallizes into the tetragonal CeFeSi-type structure and is considered as a non-magnetic heavy-fermion system [4], has been recently studied. It has been discovered that CeRuSiH retains the same crystalline structure, although the insertion of hydrogen involves an anisotropic expansion of the unit cell volume, inducing magnetic order [5]. Indeed, CeRuSiH exhibits a complex magnetic phase diagram in that two antiferromagnetic transitions at $T_{N1} = 7.9$ K and $T_{N2} = 3.1$ K have been observed. Furthermore, a metamagnetic double transition was evidenced at low temperatures [5]. These results, supported by the theoretical analysis of the chemical bonding properties [6], suggested that in CeRuSiH, bonding effects of hydrogen do not dominate over cell expansion, which favours the magnetic order. To gain further insight into this issue, in the present work, we study the effect of hydrostatic pressure on this hydride.

2. Experimental details

The CeRuSiH sample was prepared by heating an annealed ingot of CeRuSi at 523 K under vacuum for 12 hours and then exposed for 2 days under 4 MPa of hydrogen gas at the same temperature. For more details of sample preparation, see reference [5].

Magnetization measurements have been performed in a commercial (Quantum Design) SQUID magnetometer using a miniature piston-cylinder CuBe pressure cell. A small piece of Sn metal placed inside the cell was used as a manometer. The empty pressure cell was measured separately and the signal corrected accordingly. The magnetization has been measured in hydrostatic pressures up to $P = 10.3$ kbar (at ~ 3.5 K) at temperatures in the range 2-15 K and in magnetic fields up to 50 kOe.

For the zero-field-cooled (ZFC) measurements, the sample was cooled down to 2 K in the absence of an external magnetic field and magnetization measured in a constant magnetic field during the warming run of the experiments. For the field-cooled (FC) measurements, the sample was cooled from 15 K down to 2 K in the presence of the same constant magnetic field, which was used in the previous warm up scan.

3. Results and Discussion

The ZFC and FC susceptibility curves at a field of 2 kOe and the FC curves at the fields of 10 kOe and 40 kOe, for different pressures, are depicted in the figures 1a, 1b and 1c, respectively, while the field dependence of magnetization at various fixed pressures is displayed in figure 2. At 2 kOe and $P = 0$ kbar, the susceptibility shows two maxima characteristic of the two magnetic transitions, defined by $T_{N1} = 7.9$ K and $T_{N2} = 3.1$ K. Below T_{N2} , an irreversibility in the ZFC-FC curves appears which reflects the existence of some ferromagnetic component. The effects of pressure are: 1) to decrease the susceptibility (except in the low temperature range); 2) shift the two magnetic transitions to higher temperatures, and 3) weaken the irreversibility. The effects of pressure at 10 kOe (figure 1b) and 40 kOe (figure 1c) are similar to those observed at 2 kOe. The most significant difference is a strong reduction in the susceptibility observed below T_{N2} in the 10 kOe curves, due to the complex magnetic phase diagram.

To monitor the evolution of the transition temperatures T_{N1} and T_{N2} with pressure, these temperatures have been accurately determined from the temperature derivative of susceptibility ($d\chi/dT$) following the same criteria as in reference [5]. The results are depicted in figure 3. A linear behaviour is observed, with a similar pressure dependence for the different fields. In particular, at 2 kOe, T_{N1} increases from 7.9 K at $P = 0$ kbar to 9.4 K at $P = 10.3$ kbar, yielding a pressure coefficient $dT_{N1}/dP = + 0.14$ K/kbar. This value is larger than that found in the antiferromagnetic stannide CeNiSnH: $dT_N/dP = + 0.022$ K/kbar [7], but similar to that found in the antiferromagnetic CeNiGe₃: $dT_N/dP = + 0.15$ K/kbar [8], or the ferromagnetic CePbSb: $dT_C/dP = + 0.16$ K/kbar [9].

This pressure dependence is understood within the framework of the Doniach's phase diagram, which describes the competition between the Kondo effect and the indirect magnetic RKKY interactions as a function of the coupling J_{cf} between $4f(\text{Ce})$ and conduction electrons [10]. Initially, the increase of volume induced by the hydrogen insertion decreases drastically J_{cf} , so that the CeRuSiH compound falls in the left part of the Doniach diagram. The application of pressure induces the opposite effect, increases continuously J_{cf} and, in this part of diagram, results in an enhancement of the RKKY interactions compared to the Kondo effect, increasing the order temperature. In other words, the pressure shifts the properties of the compound from the left towards the center of the Doniach diagram.

Each of the $M(H)$ isobars taken at $T = 2$ K and $T = 5$ K, and shown in figure 2, has been measured first while increasing the field up to $H = 50$ kOe and then, decreasing the field down to $H = 0$ kOe. The curve corresponding to $T = 2$ K and $P = 0$ kbar (Figure 2a) shows two metamagnetic transitions at $H_{C1} = 6.7$ kOe and $H_{C2} \approx 48$ kOe. Additionally, the

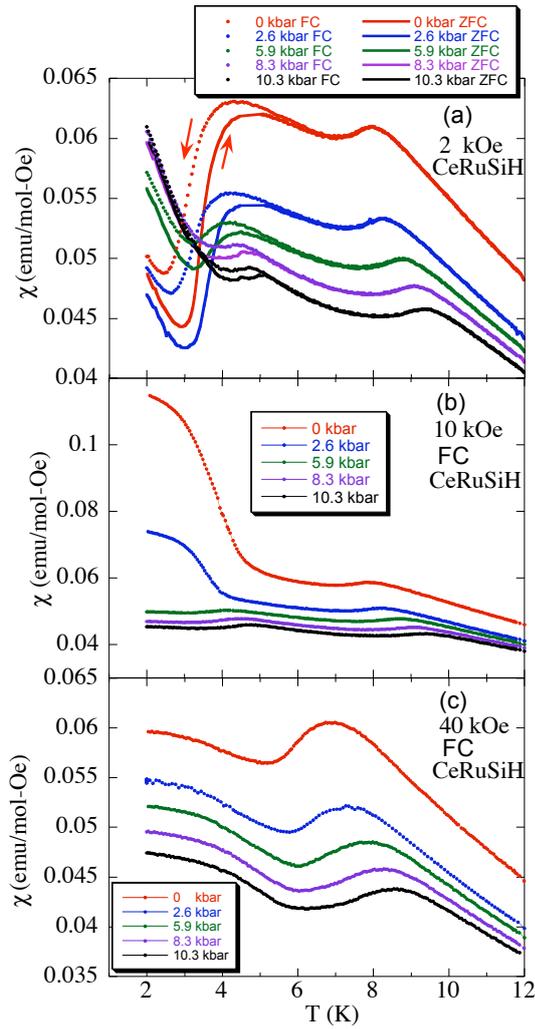


Figure 1. Temperature dependence of the susceptibility of CeRuSiH measured under various external pressures and for magnetic fields : (a) $H = 2$ kOe, (b) $H = 10$ kOe and (c) $H = 40$ kOe.

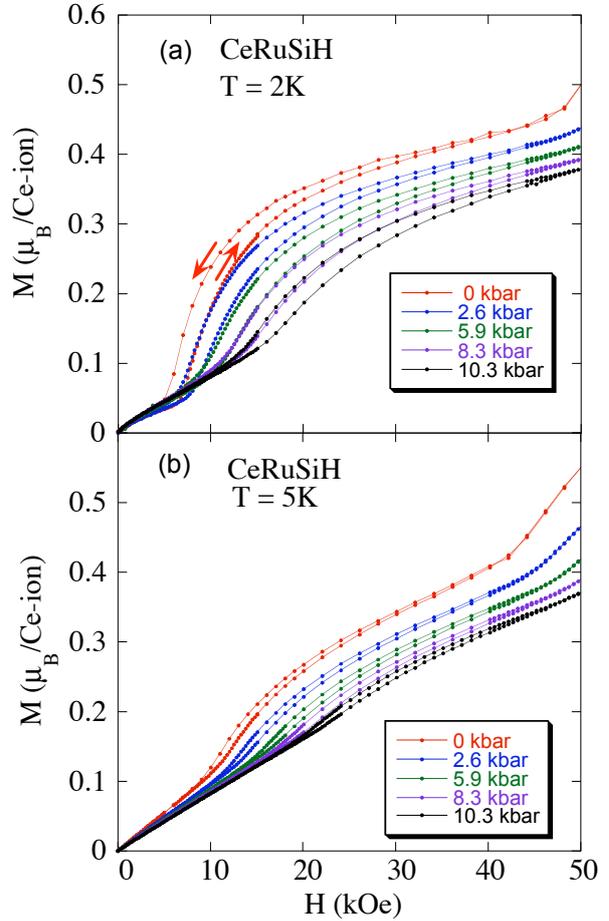


Figure 2. Field dependence of the magnetization of CeRuSiH measured under various external pressures and for temperatures (a) $T = 2$ K and (b) $T = 5$ K.

magnetization exhibits a suggestive irreversibility between H_{C1} and H_{C2} , which had not been previously detected [5]. The effect of pressure is to shift the critical fields H_{C1} and H_{C2} to higher values, keeping nearly constant the irreversibility. At $T = 5$ K and $P = 0$ kbar (Figure 2b), the value of H_{C1} is higher than the observed at $T = 2$ K whereas the value of H_{C2} is lower; a small irreversibility between H_{C1} and H_{C2} still remains. At this temperature, the pressure gives rise to an evolution of the critical fields similar to that described at $T = 2$ K. The pressure dependencies of H_{C1} and H_{C2} are depicted in figure 4. In particular, for H_{C1} we represent separately the values obtained while increasing and decreasing the magnetic field. In all the cases, the critical fields increase nearly linearly with pressure, with a pressure coefficient $dH_C/dT \sim 0.90$ kOe/kbar, which, like the Néel temperatures, is about six times larger than that found in CeNiSnH ($dH_C/dT \sim 0.15$ kOe/kbar). These correlations between the critical field and the ordering temperature suggest that the enhancement in the RKKY interactions with pressure, as previously commented, is also responsible for the increase of the critical fields.

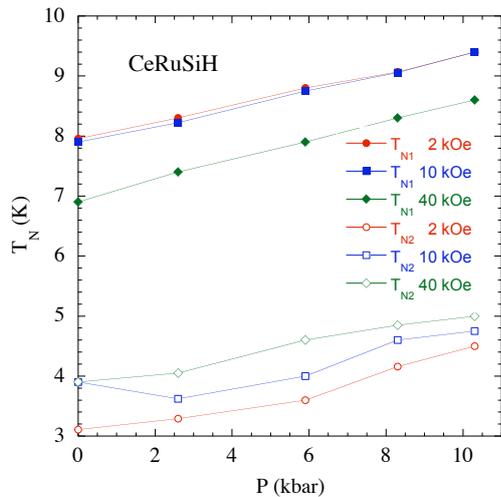


Figure 3. Pressure dependence of T_{N1} and T_{N2} ordering temperatures for CeRuSiH obtained at different magnetic fields.

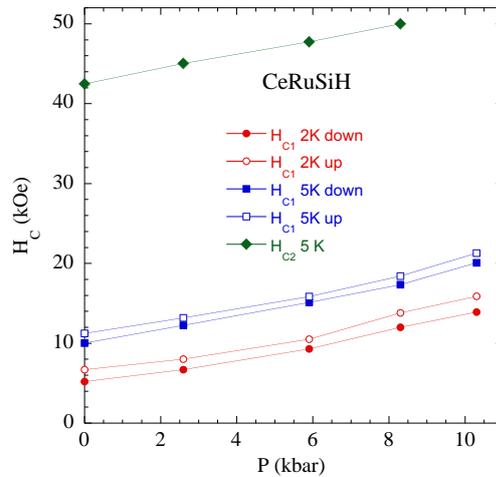


Figure 4. Pressure dependence of the critical fields H_{C1} and H_{C2} obtained at $T = 2$ K and $T = 5$ K.

4. Conclusion

The influence of H insertion on CeRuSi is to establish a 3+ valence on Ce ions, situating the compound in the left part of the Doniach diagram. The application of pressure increases the hybridization of the $4f(\text{Ce})$ electrons with the conduction band shifting the compound towards the centre of the diagram and subsequently increasing the ordering temperature. Therefore, pressure has an effect opposite to that of hydrogen insertion, confirming that bonding effect plays a minor role, compared to volume effects, during hydrogenation of this silicide. Finally, it is expected that a further increase in pressure will make T_N pass through a maximum and then tends to zero, thereby reverting the effects of hydrogenation.

Acknowledgments

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