Comparison of a Shastry-Sutherland lattice stability in Ce$_2$Pd$_2$Sn as a function of field and doping

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Abstract. A comparative study of the stability of a Shastry-Sutherland lattice (SSL) in Ce$_2$Pd$_2$Sn under different physical conditions is presented. Applied magnetic field suppresses the SSL of stoichiometric Ce$_2$Pd$_2$Sn in a magnetic critical point at $T_{cr}(B) = 4.2$ K and $B_{cr} = 0.13$ T, whereas 25% of Ni does it at $T_{cr}(Ni) = 3.4$ K. Electronic concentration variation, driven by the increase of Pd (holes) concentration in Ce$_2$Pd$_{2+y}$In$_{1-y}$, decreases the magnetic transition down to $T_M = 2.8$ K in the limit of the alloy solubility, i.e. $y = 0.4$. The existence of a $M(B)$ plateau in SSL predicted by theory and the crossing of those isotherms, previously observed in the model compound SrCu$_2$(BO$_3$)$_2$ are analyzed.

1. Introduction

The Shastry-Sutherland lattice (SSL) can be described as a 2D square lattice built up from a network of dimers disposed orthogonally to each other [1]. Those dimers form from an antiferromagnetic AF coupling $J$ between magnetic nearest-neighbors $nn$ atoms, which interact among them through a $J'$ coupling. Such a configuration is topologically equivalent to the 2D square lattice Heisenberg model [1] which has an exact dimer ground state. The model compound for this type of magnetic lattice is SrCu$_2$(BO$_3$)$_2$ [2], though it was recently found to form also in some intermetallic compounds with the Mo$_2$FeB$_2$-type of structure [3].

In this contribution we present a comparative study of the SSL stability in Ce$_2$T$_2$X intermetallic compounds, where T = transition metal and X = p-metal. For that purpose we have applied different control parameters, such as: i) magnetic field, ii) T = transition-metal substitution to induce a structural pressure and iii) the variation of electronic concentration (i.e. the chemical potential).

2. Experimental and results

The studied samples are polycrystalline, prepared by arc melting the appropriate amount of components. The respective annealing procedures and X-ray characterization were described elsewhere, Ref.[4] for Ce$_2$Pd$_2$Sn and [5] for Ce$_2$Pd$_{2+y}$In$_{1-y}$ family. Specific heat was measured using the heat pulse technique in a semi-adiabatic He-3 calorimeter in the range between 0.5 and 20K, at zero and applied magnetic field up to 4T. DC-magnetization measurements were carried out using a standard SQUID magnetometer operating between 2 and 300K, and as a function of field up to 5T.
3. Magnetic field effect in Ce$_2$Pd$_2$Sn

The effect of magnetic field was investigated in stoichiometric and alloyed samples. However, for simplicity we only analyze here the results on stoichiometric Ce$_2$Pd$_2$Sn which is the best example showing the main characteristics of SSL under magnetic field. In this compound, the formation of a SSL in Ce$_2$Pd$_2$Sn was observed within a limited range of temperature, between an (second order) AF transition at $T_M = 4.9$ K and a (first order) ferromagnetic FM one at $T_C = 2.1$ K [6], having as the upper limit a correlated paramagnetic phase and a FM one at low temperature. In Ce$_2$Pd$_2$Sn this exotic phase builds up from FM-dimers formed by Ce $nn$ atoms, which are coupled by $J$. An AF $J'$ coupling between those dimers drives the formation of the SSL, realized as a quasi 2D square lattice of effective spin $S_{eff}$ = 1 below $T_M = 4.9$ K. Since the SSL only holds within a short range of temperature it is strongly affected by external magnetic field, including its suppression at a magnetic critical point: $B_{cr} = 0.13$ T and $T_{cr} = 4.1$ K [7].

\[ \frac{\delta M}{\delta B} \approx 1 \text{ value} \]

Isothermal $M(B)$ measurements show some peculiar features characteristic of the SSL, which were previously observed in the model compound SrCu$_2$(BO$_3$)$_2$ [2, 8], such as quantized magnetization plateaux at fractional values of the saturation moment $M_{sat}$ (c.f. 1/4 and 1/8 $M_{sat}$) and the crossing of magnetization isotherms as a function of $B$ at certain temperatures. One of those plateaux is revealed incipiently in Ce$_2$Pd$_2$Sn as a satellite maximum in the $\frac{\delta M}{\delta B} \vert_{T}$ derivative, see Fig. 1. In the magnetic phase diagram presented in Fig. 2, the low temperature phase boundary (related to $T_C$) was extracted from the maximum of $\frac{\delta M}{\delta B} \vert_{T}$ for $2 \leq T \leq 5$ K (only shown for $T > 3.9$ K in Fig. 2). The upper phase boundary (related to $T_M(B)$) was extracted from the cusp of $M(T)$ at $T_M(B)$, the maximum of $\frac{\delta M}{\delta B} \vert_{T}$ and its satellite .

\[ \frac{\delta M}{\delta B} \triangle \]

\[ \text{SSL} \]

\[ \text{Correlated Paramagnetism} \]

\[ \text{FM} \]

\[ \text{1st. max} \]

\[ \text{2nd. max} \]

**Figure 1.** Detail of the $M(B)$ derivative of Ce$_2$Pd$_2$Sn within the $3.9 \leq T \leq 5$ K range showing the satellite maximum (dashed curve) around the critical field.

**Figure 2.** Phase diagram of Ce$_2$Pd$_2$Sn. The phase boundaries are extracted from the cusp of $M(T)$ at $T_M(B)$, the maximum of $\frac{\delta M}{\delta B} \vert_{T}$ and its satellite .
4. Structural pressure in Ni doped Ce$_2$(Pd$_{1-x}$Ni$_x$)$_2$Sn

The structural pressure was induced by doping the Pd lattice with smaller isoelectronic Ni atoms. In this case a weakening of the Ce magnetic moment is expected due to the Kondo screening of the 4f Ce states. In Ce$_2$(Pd$_{1-x}$Ni$_x$)$_2$Sn that effect is observed as a reduction of the upper ordering temperature $T_M$ as a function of Ni concentration $x(Ni)$, see Fig. 3. On the contrary, the lower transition $T_C$ to the FM phase increases with $x(Ni)$ as a consequence of the reduction of the temperature range of the SSL stability. This behavior can be explained by the weakening of the $J$ and $J'$ couplings due to the increasing Kondo screening. Those couplings may be eventually overcome by the inter-Ce planes coupling in the ‘c’ crystalline direction $J_C$, which for $x \approx 0.25$ practically inhibits the formation of the SSL phase.

The arising Kondo screening is confirmed by i) the increasing $\gamma$ coefficient (extracted from a $C_m/T(T \to 0)$ extrapolation), which increases proportionally to $x(Ni)$ and ii) the weakening of the saturation magnetic moment at high field (not shown). In Fig. 4 we present Ni dependent phase diagram which extrapolates to a critical concentration $x_{cr}(Ni) \approx 0.27$.

![Figure 3](image1.png)

**Figure 3.** Thermal dependence of specific heat showing upper ($T_M$) and lower ($T_C$) transitions. The magnetic contribution $C_m$ was evaluated after La$_2$Pd$_2$Sn phonon subtraction.

![Figure 4](image2.png)

**Figure 4.** Ni doping dependencies of $T_M$ and $T_C$, which extrapolate to a critical point at $T_{cr}(Ni) \approx 3.5$ K for $x_{cr}(Ni) \approx 0.27$, where the SSL is suppressed.

5. Electron concentration effect in Ce$_2$Pd$_{2+y}$In$_{1-y}$

The change of electronic concentration and consequently the chemical potential, is realized in the isotypic compound Ce$_2$Pd$_{2+y}$In$_{1-y}$. In that compound one takes profit of its extended range of solubility, between 0.1 \( \leq y \leq 0.4 \) [5], where the actual Ce atomic concentration is 1.95 instead of 2. The electronic variation is induced by enriching the Pd (holes) concentration and by the replacement of Sn[5s$^2$5p$^2$] by In[5s$^2$5p$^1$].

In Fig. 5 we present the magnetic contribution to the specific heat which shows that the transition $T_M$ decreases upon increasing $y(Pd)$. In these system no $T_C$ transition is observed but a kink (at $T = T^*$) in the $y(Pd) = 2.20$ and 0.25 alloys, which transforms into a shoulder for higher Pd concentrations. Coincidentally, only the former alloys show the SSL signs, i.e. the crossing of their $M(B)$ isotherms and a satellite maximum in $\partial S/\partial B \mid_T = 0$ like in Ce$_2$Pd$_2$Sn. Those properties vanish for $y(Pd) = 2.35$ and 0.24 concentrations indicating that the SSL is strongly affected by the Pd/In modification which also increases atomic disorder.
Figure 5. Magnetic specific heat dependence on temperature and \(y(Pd)\) concentration. Basal plane: projection of the \(y(Pd)\) dependence of \(T_M\) and \(T^*\), see the text.

Figure 6. (a) Electronic, \(y(Pd)\) holes, dependence of the \(T_M\) phase boundary and \(T^*\). (b) Field dependence for the richest \(y(Pd) = 0.24\) alloy. Dot line: extrapolation of \(T^*\) to the critical field.

6. Conclusions

We have compared the stability of the SSL in the Ce\(_2\)T\(_2\)X family of compounds (\(T=\) Pd, Ni and X= Sn, In) driving different control parameters, c.f. magnetic field, structural pressure and electron concentration. We find that the SSL is suppressed in stoichiometric Ce\(_2\)Pd\(_2\)Sn by a moderate field \((B_{cr} = 0.13\) T), by arising the Kondo screening of Ce moments with \(x_{cr}(Ni) \approx 0.27\) doping, and by increasing electron (holes) concentration up to \(y(Pd) = 2.25\) in Ce\(_{1.95}\)Pd\(_{2+y}\)In\(_{1-y}\). Comparatively, a stronger magnetic field is required to induce FM polarization in indeede compounds than in the stannides, indicating a different relation between the on-plane \(J\) and \(J'\) coupling parameters strength respect to the inter-plane \(J_C\) one.

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