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Supplemental Materials for "Anisotropic Magnetic Field–Temperature Phase Diagrams in CeCoSi"

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1. Determination of T_0 and T_{s1} from $d^2(M(T)/H)/dT^2$

The temperature dependence of M/H in CeCoSi (sample #2) for $H \parallel [100]$ and [110] between 1 and 7 T are shown in Figs. S1(a) and S1(b), respectively. In addition, their second derivative [$= d^2(M(T)/H)/dT^2$] for $H \parallel [100]$ and [110] are shown in Figs. S1(c) and S1(d), respectively. For $H \parallel [100]$, the $d^2(M(T)/H)/dT^2$ curves show the maximum at T_0 and the minimum at T_{s1}. The maximum and minimum in $d^2(M(T)/H)/dT^2$ below 2 T should be interpreted as the apparent T_0 and T_{s1}, respectively. On the other hand, the $d^2(M(T)/H)/dT^2$ curves for $H \parallel [110]$ show the maximum at T_0, but no anomaly corresponding to T_{s1} can be observed.

Figure S2 shows the temperature dependence of $d^2(M/H)/dT^2$ in CeCoSi (sample #1) under $H \parallel [001]$ below 7 T. The $d^2(M(T)/H)/dT^2$ curves show the maximum at T_0, while no anomaly corresponding to the T_{s1} transition is observed. Here, the raw $M(T)/H$ data are taken from our previous paper.[15)]

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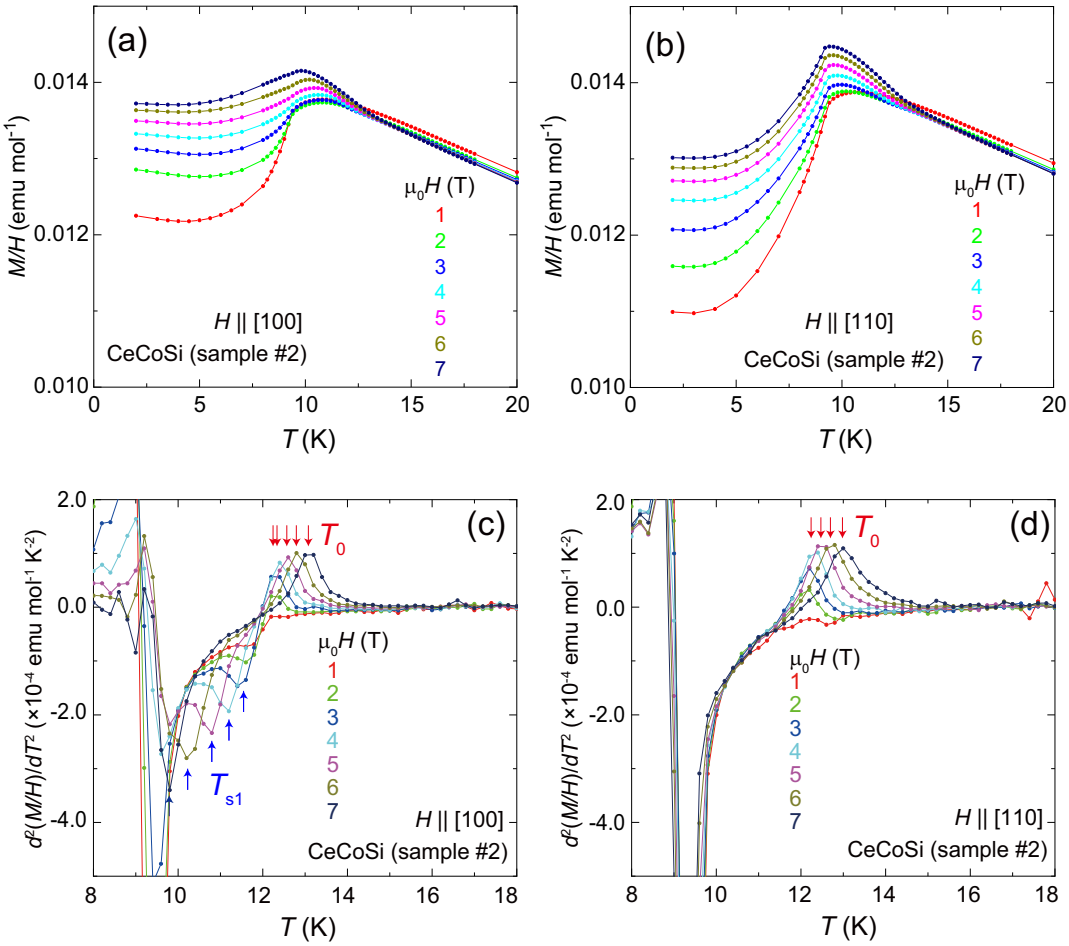

Fig. S1. (Color online) Temperature dependence of M/H in CeCoSi (sample #2) for H || (a) [100] and (b) [110] between 1 and 7 T. Their second derivative with respect to temperature [$= d^2(M(T)/H)/dT^2$] are shown in (c) and (d), respectively.

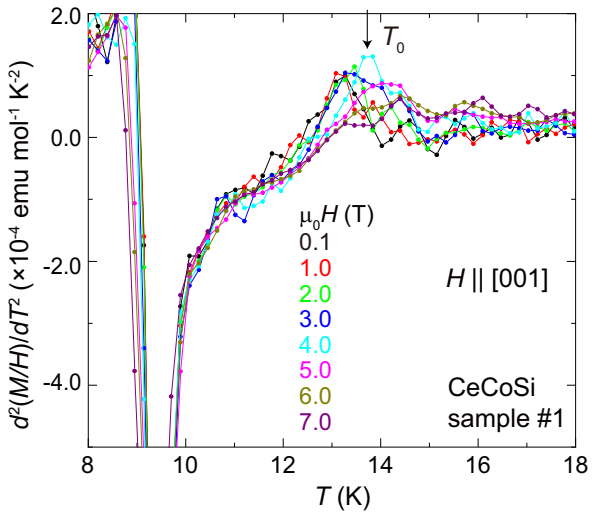

Fig. S2. (Color online) Second derivative of $M(T)/H$ in CeCoSi (sample #1) under H || [001] below 7 T.

2. Specific heat data under H || [100] and [110]

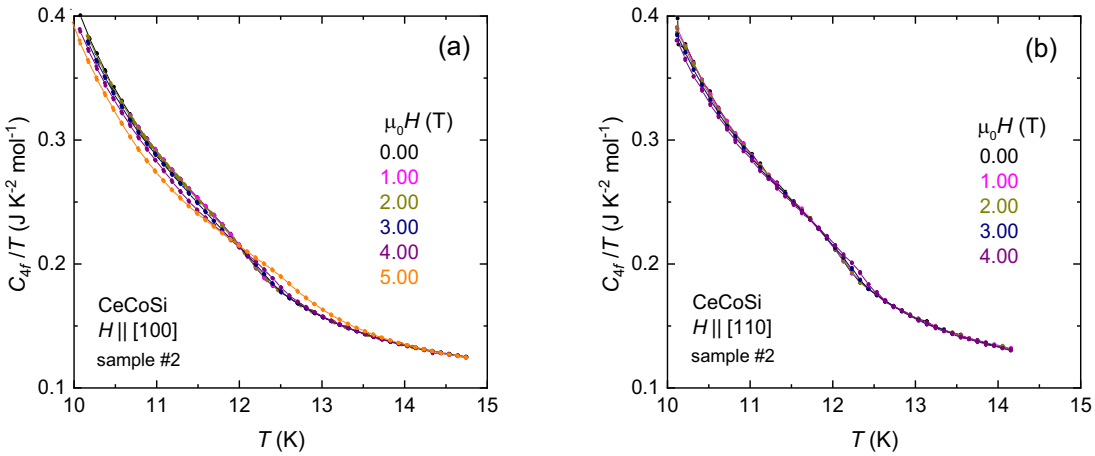

Fig. S3. (Color online) Temperature dependence of C_{4f}/T in CeCoSi (sample #2) under H || (a) [100] and (b) [110].

The temperature dependence of C_{4f}/T in CeCoSi (sample #2) under H || [100] and [110] are shown in Figs. S3(a) and S3(b), respectively. Here, C_{4f} is the contribution of $4f$ electrons to the specific heat, estimated by subtracting the C of LaCoSi from the C of the present measured sample.[12] The first derivative data [= $d(C_{4f}/T)/dT$] for H || [100] and [110] are shown in Figs. S4(a) and S4(b), respectively. The $d(C_{4f}/T)/dT$ curves show the minimum at T_0 and T_{s1}. The error for each transition point was determined by the full width at half maximum of the peak anomaly in $d(C_{4f}/T)/dT$. At 3 T for H || [100], the T_{s1} determined from the $M(T)$ measurements is shown for comparison.

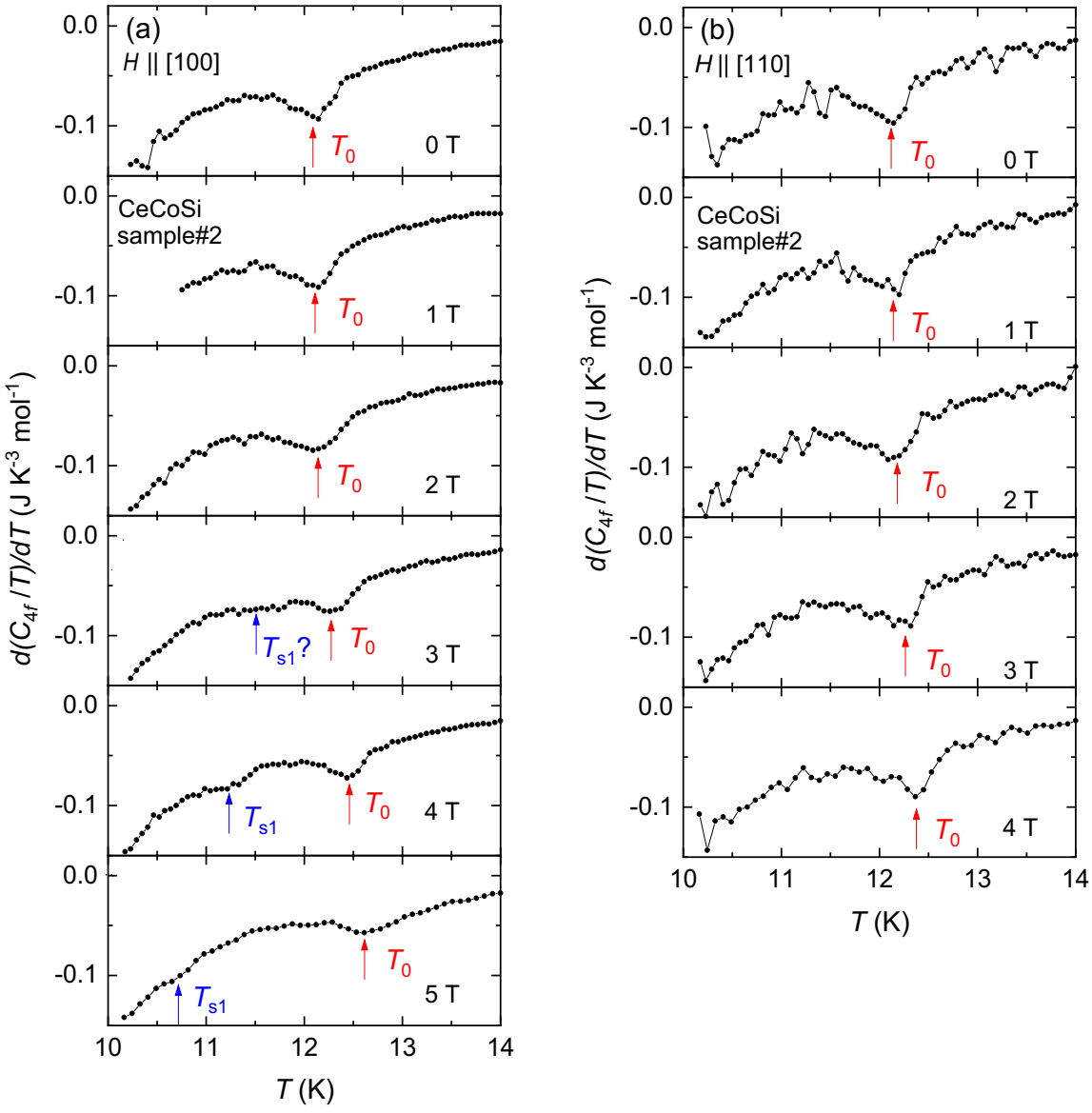

Fig. S4. (Color online) First derivative of $C_{4f}(T)/T$ in CeCoSi (sample #2) under H || (a) [100] and (b) [110]. They were obtained by differentiating the interpolated data of $C_{4f}(T)/T$.

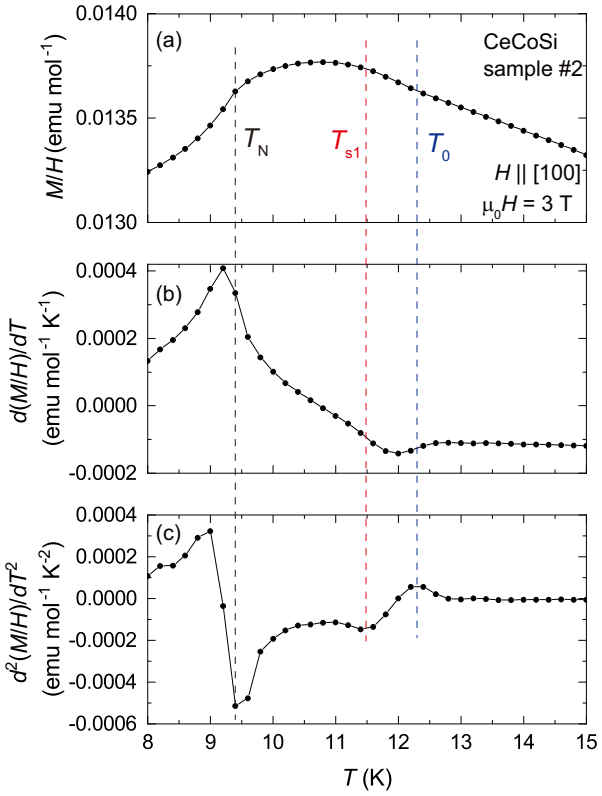

Fig. S5. (Color online) (a) $M(T)/H$, (b) first derivative of $M(T)/H$, and (c) second derivative of $M(T)/H$ in CeCoSi (sample #2) under H || [100] at 3 T.

3. Interpretation of the T_{s1} and T_0 phase boundaries by the Ehrenfest relation

Figures S5(a)-(c) show the temperature dependence of M/H, the first derivative of $M(T)/H$ [= $d(M(T)/H)/dT$], and the second derivative of $M(T)/H$ [= $d^2(M(T)/H)/dT^2$], respectively, in CeCoSi (sample #2) under H || [100] at 3 T. The increase in T_0 and the decrease in T_{s1} by applying H can be explained by the observed behavior in $M(T)$ and $C(T)$ at each transition temperature through the Ehrenfest relation for a second order phase transition:

$$\Delta\left(\frac{\partial M}{\partial T}\right) = -\frac{dT_x}{dH}\Delta\left(\frac{C}{T}\right). \quad (1)$$

Here, ΔX means a discontinuity of the quantity X across the second-order phase boundary and T_x is a phase transition temperature.[19] When the system enters an ordered phase from the high temperature side, $\Delta(C/T)$ is always positive. Thus the decrease in $d(M(T)/H)/dT$ at T_0, which corresponds to the maximum in the second derivative of $M(T)/H$, means the increase of T_0 with increasing H, while the increase in $d(M(T)/H)/dT$ at T_{s1} the decrease of T_{s1}.

4. Simulation of the superposition of maximum and minimum

Figure S6 shows the simulation result of the superposition of maximum and minimum. Here, the Gaussian function is assumed. The purple curves represent the results of the superposition of the red- and blue-dashed curves.

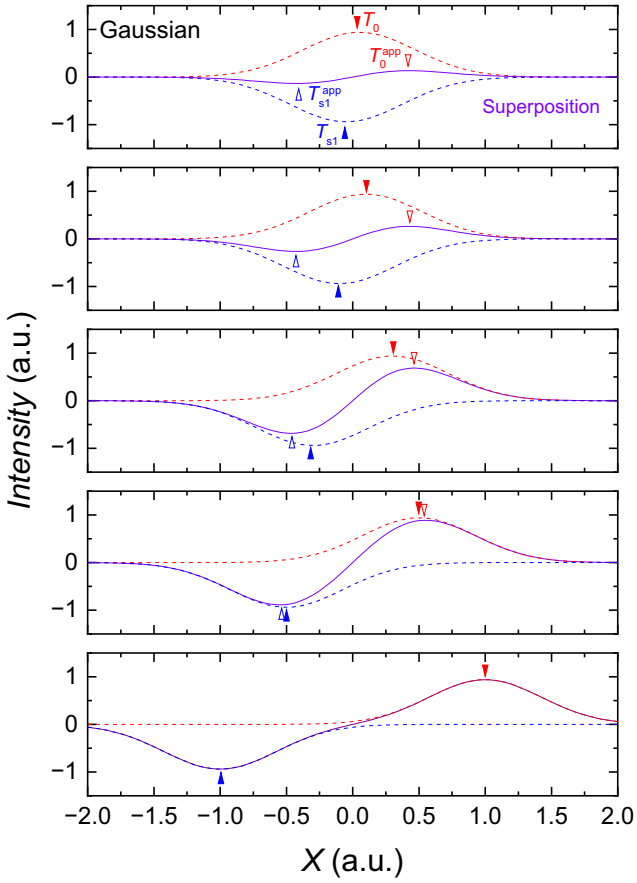

Fig. S6. (Color online) Simulation of the superposition of maximum and minimum. The purple curves represent the results of the superposition of the red- and blue-dashed curves. The closed and open triangles indicate the true and apparent extrema, respectively, such as T_0 and T_0^{app}.

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