Supplemental Materials for "Magnetic field angle dependence of the magnetic phase diagram in UNi_4B"

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I. CORRECTION OF LINEAR TIME DEPENDENCE IN MAGNETOSTRICTION

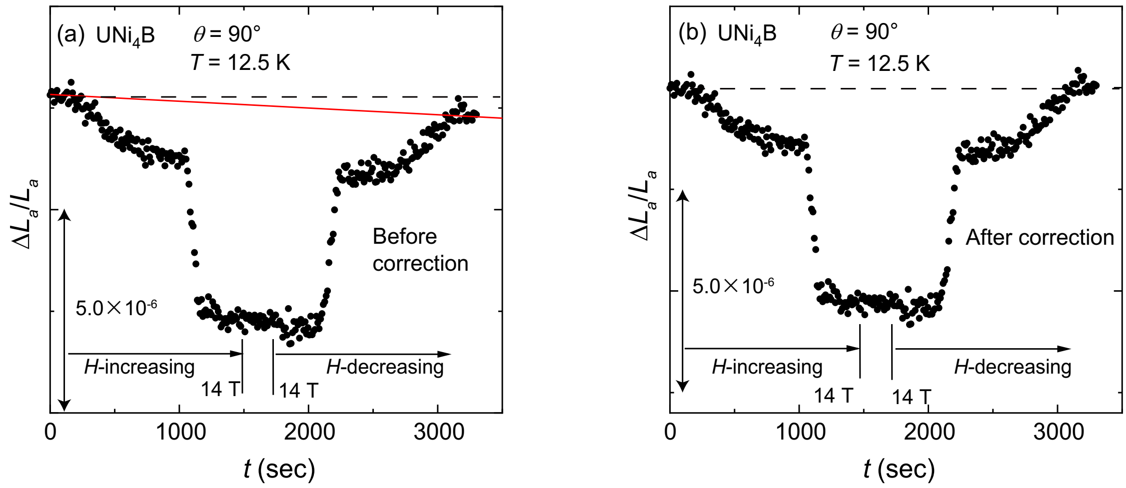

FIG. S1. (Color online) Time dependence of $\Delta L_a/L_a$ during the magnetostriction measurements at 12.5 K for $\theta = 90°$ in UNi_4B. (a) the data before removing the linear time dependence, as represented by the red solid line, and (b) the data after removing the linear time dependence.

We show the typical example of the time dependence of $\Delta L_a/L_a$ in UNi_4B during the magnetostriction measurements in Fig. S1. As can be seen in (a), the data contains a linear time-dependent component that is likely attributed to extrinsic factors. The data after removing the linear time-dependent component is shown in (b).

II. INFLUENCE OF JOULE HEATING INDUCED BY APPLIED CURRENT

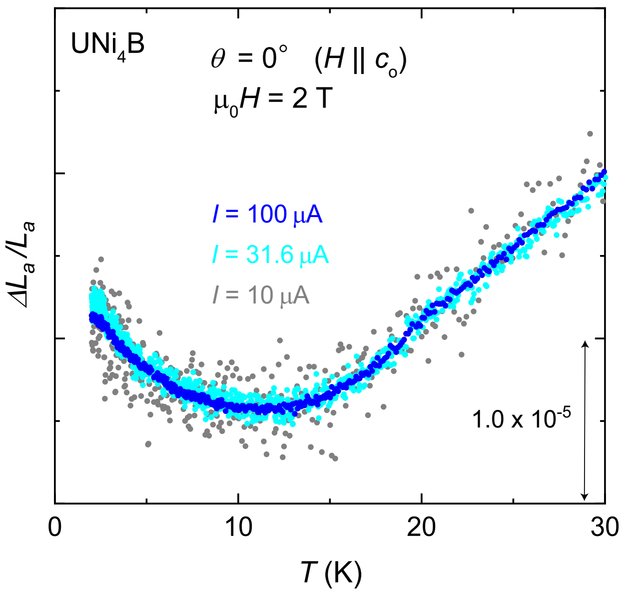

FIG. S2. (Color online) $\Delta L_a(T)/L_a$ of UNi_4B at 2 T for $\theta = 0°$ obtained under different electric currents applied to the circuit.

III. THERMAL EXPANSION COEFFICIENT AT ZERO MAGNETIC FIELD

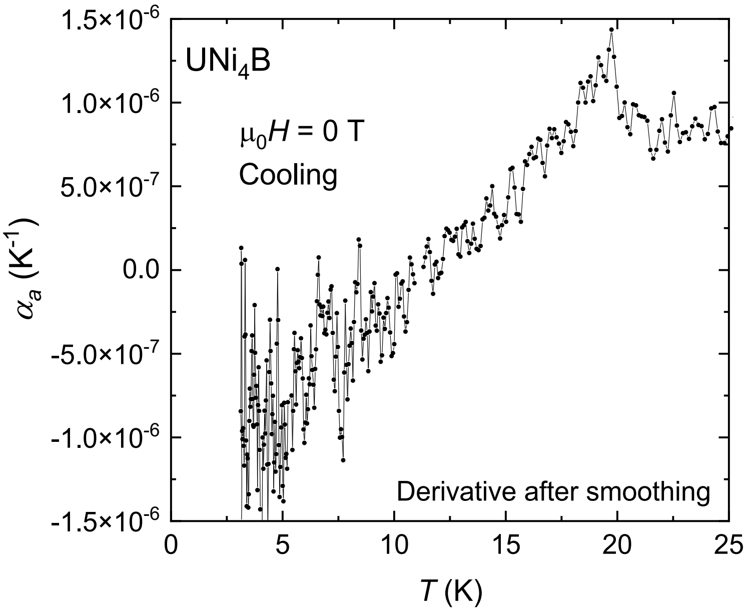

FIG. S3. Temperature dependence of α_a in UNi_4B at zero magnetic field.

We show the temperature dependence of thermal expansion coefficient along the $a_{\rm o}$ axis ($= \alpha_a$) in UNi_4B measured at zero magnetic field. The $\alpha_a(T)$ was obtained by differentiating the $\Delta L_a(T)/L_a$ after smoothing. The magnitude of jump at $T_{\rm N}$ observed in our data is comparable to that in the previous data reported by Mentink et al. [21] On the other hand, our data show larger negative values below $\sim$ 10 K than those reported by Mentink et al. [21], which may be attributed to the reduced accuracy in the absolute values measured by the active-dummy method in the low temperature region of a few Kelvin. This difference in α_a below $\sim$10 K is not significant in this study, which aims to investigate the anisotropy of transition points in the magnetic phase diagram.

IV. $\Delta L_a(T)/L_a$ ON COOLING AND HEATING PROCESSES

Figures S4 show the $\Delta L_a(T)/L_a$ curves in UNi_4B during the field-cooling (FC) and subsequent heating processes at (a) $\mu_0 H = 8$ and (b) 12 T for $\theta = 0°$, and at (c) 8 and (d) 12 T for $\theta = 60°$. The transition temperatures were estimated based on the results of both the cooling and heating processes, as indicated in these figures. Note that a larger difference in $\Delta L_a(T)/L_a$ for $\theta = 60°$ between the cooling and heating processes has been observed in the data for all intermediate angles, which may be caused by the use of the plastic sample holder.

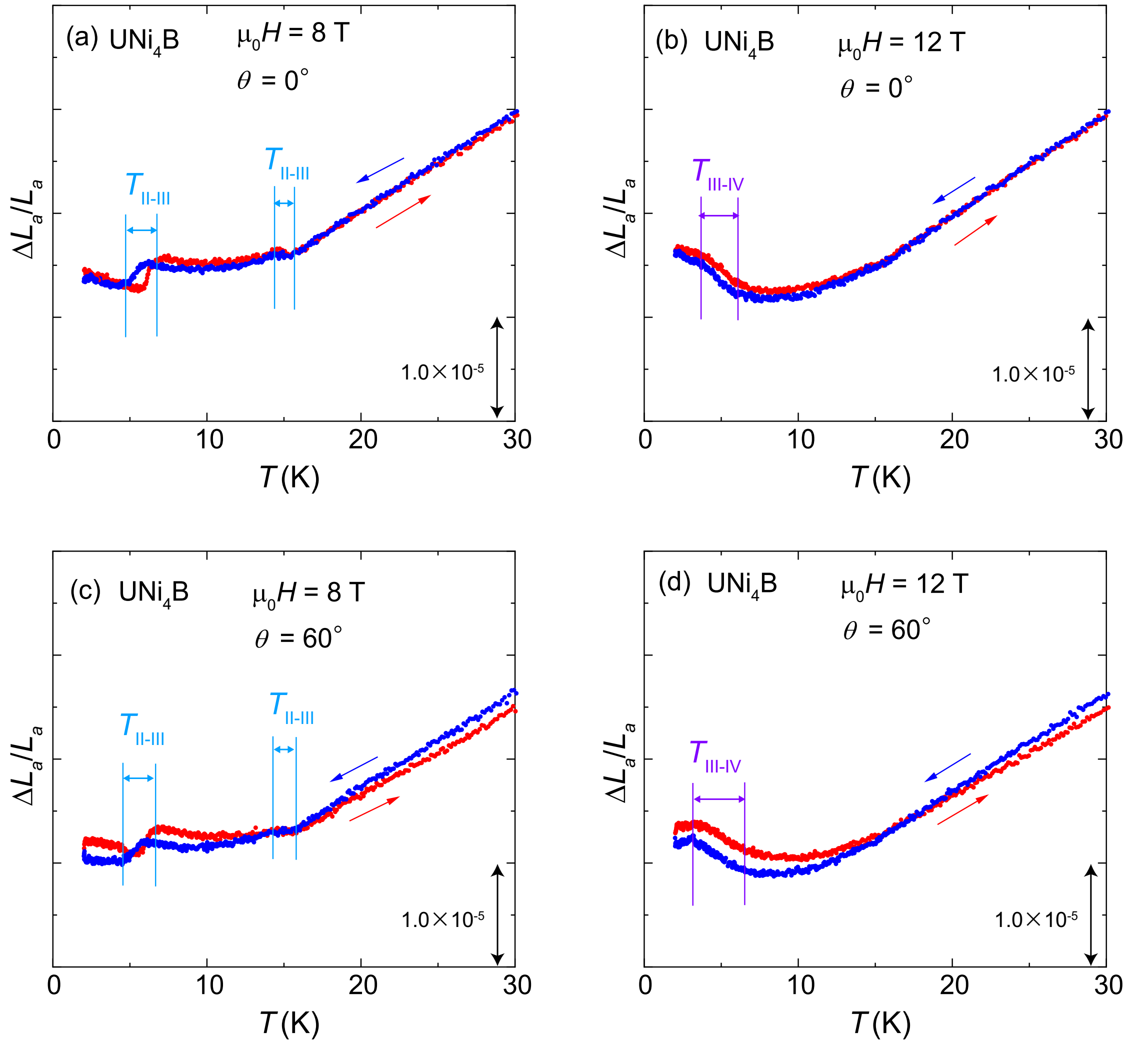

FIG. S4. (Color online) Temperature dependence of $\Delta L_a/L_a$ during the FC and subsequent heating processes in UNi_4B at $\mu_0 H =$ (a) 8 and (b) 12 T for $\theta = 0°$, and at (c) 8 and (d) 12 T for $\theta = 60°$.

V. COMPARISON OF PREVIOUSLY REPORTED H-T PHASE DIAGRAMS

Figures S5(a) and S5(b) show the previously reported H-T phase diagrams in UNi_4B for $H \parallel$ (a) a_h and (b) b_h in the hexagonal structure, respectively. The transition points determined by Mentink et al. and by Yanagisawa et al. are represented by open and closed circles, respectively. [17, 18] Note that Yanagisawa's data are plotted by swapping the a_h and b_h axes in these figures, which show good agreement with those determined from the Mentink's study and the present thermal expansion study [see Figs. 2(a) and 2(h) in the main text]. In addition, the presence of hysteresis at H_{1b} and the absence of hysteresis at H_{2b} in the H dependence of C_{66} of ultrasound measurements for $H \parallel b_h$ (= $[01\bar{1}0]$) [17] are also consistent with our $\Delta L_a(H)/L_a$ results for $\theta = 0°$ and $\theta = 60°$ [see Figs. 1(c) and 3(a) in the main text].

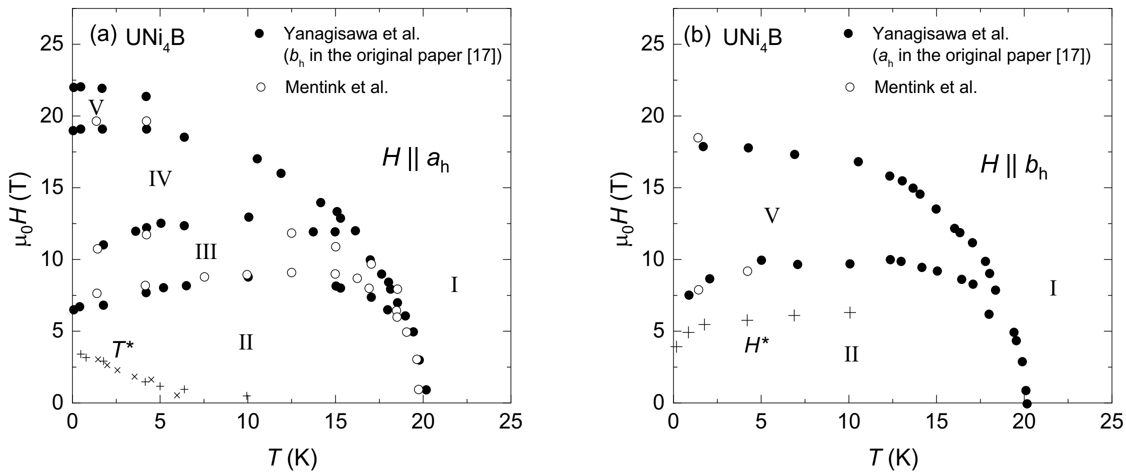

FIG. S5. Previously reported H-T phase diagrams of UNi_4B for $H \parallel$ (a) a_h and (b) b_h. The transition points determined by Mentink et al. and by Yanagisawa et al. are represented by open and closed circles, respectively. [17, 18] Here, Yanagisawa's data are plotted by swapping the a_h and b_h axes.

[17] T. Yanagisawa, H. Matsumori, H. Saito, H. Hidaka, H. Amitsuka, S. Nakamura, S. Awaji, D. I. Gorbunov, S. Zherlitsyn, J. Wosnitza, K. Uhlířová, M. Vališka, and V. Sechovský, Electric quadrupolar contributions in the magnetic phases of UNi_4B, Phys. Rev. Lett. **126**, 157201 (2021).

[18] S. A. M. Mentink, G. J. Nieuwenhuys, H. Nakotte, A. A. Menovsky, A. Drost, E. Frikkee, and J. A. Mydosh, Magnetization and resistivity of UNi_4B in high magnetic fields, Phys. Rev. B **51**, 11567 (1995).

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