*Electronic supplementary information for*

# Ultrafast carrier and coherent phonon dynamics in van der Waals ferromagnet $CrI_3$

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# *Contents*

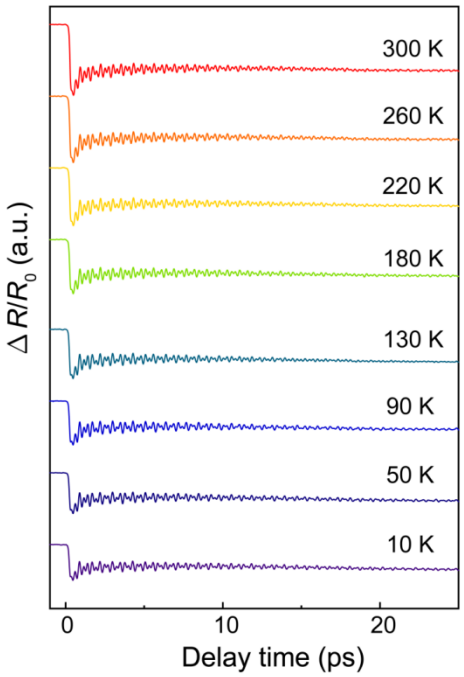


**FIG. S1.** Representative $\Delta R(t)/R_0$ signals of $CrI_3$ collected at 10-300 K. The $\Delta R(t)/R_0$ traces are shifted along the vertical axis for clarity.

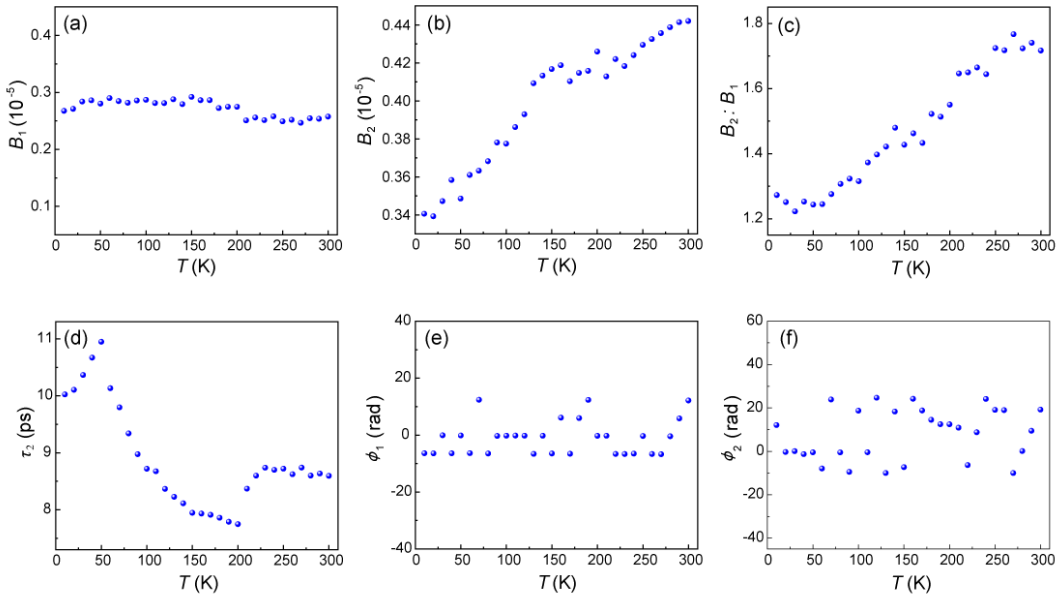


**FIG. S2.** Temperature dependence of amplitudes, (a) $B_1$ and (b) $B_2$, of the two $A_{1g}$ phonon modes. (c) The ratios ($B_2/B_1$) as a function of temperature. (d) Dephasing time $\tau_2$ of the $A_{1g}^2$ phonon mode as a function of temperature. Temperature dependence of initial phases, (e) $\phi_1$ and (f) $\phi_2$, of the two $A_{1g}$ phonon modes.

For the oscillatory part of $\Delta R(t)/R_0$ signals, the fitting with Eq. 1 also yields the amplitudes ($B_1$, $B_1$), dephasing time ($\tau_2$), and initial phases ($\phi_1$, $\phi_2$) of the two $A_{1g}$ phonon modes, as shown in **Fig. S2**. Generally, the amplitude of coherent phonons decreases with increasing temperature.[1-3] However, our data show abnormal results, although the experiments have been repeated several times. As shown in **Figs. S1(a) and 1(b)**, $B_1$ is nearly temperature independent, while $B_2$ increases with increasing temperature. This result is consistent with the reported data under left circularly polarized ($\sigma$-) pumping in a recent ultrafast study.[4] In their study, the amplitude of $A_{1g}^1$ coherent phonons is almost the same at 15 and 75 K, while the amplitude of $A_{1g}^2$ coherent phonons is much larger at 75 K than that at 15 K. The underlying reason is not known yet. **Fig. S1(d)** shows the temperature dependence of dephasing time $\tau_2$, which is similar to that of $\tau$ in Fig. 5. However, the fitting to temperature dependence of dephasing rate ($\tau_2^{-1}$) with the anharmonic phonon model is not satisfactory at 60−200 K. The dynamics of the $A_{1g}^2$ phonons is beyond our research interest.

| Parameter | Value |
|---|---|
| $\omega_0$ (rad/s) | $14.97\times10^{12}$ |
| $D$ (rad/s) | $-3\times10^{12}$ |
| $\alpha_a$ ($K^{-1}$) | $2\times10^{-6}$ |
| $\alpha_b$ ($K^{-1}$) | $2\times10^{-6}$ |
| $\alpha_c$ ($K^{-1}$) | $2\times10^{-5}$ |
| $\gamma'$ | 1.5 |

**Table S1**. Fit parameters for the $A_{1g}^{1}$ phonon mode softening in Fig. 4(a) with the anharmonicity model [Eqs (6)-(8)]. The values of linear thermal expansion coefficients ($\alpha_a$, $\alpha_b$ and $\alpha_c$) are available for $CrI_3$ from previous reports.[5]

## References


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