Supplementary materials

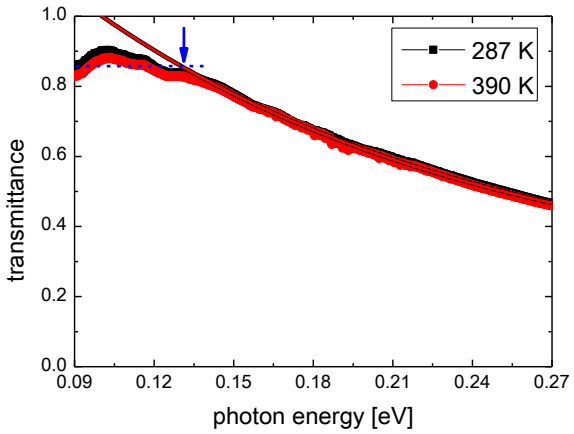

FIG. S1. IR transmittance spectra from Bi_2Te_3 at the temperatures of 287 K and 390 K. The indirect bandgap of Bi_2Te_3 is determined as 0.13 eV indicated as a blue arrow. The value of the indirect bandgap does not change in this temperature range.

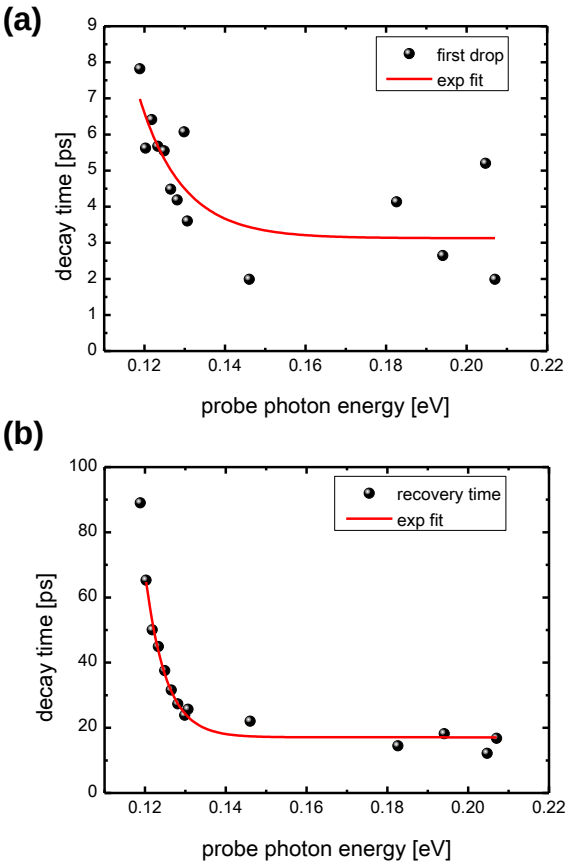

FIG. S2. The time constant of the first decrease (a) and recovery (b) in the mid-infrared probe reflectance spectroscopy.

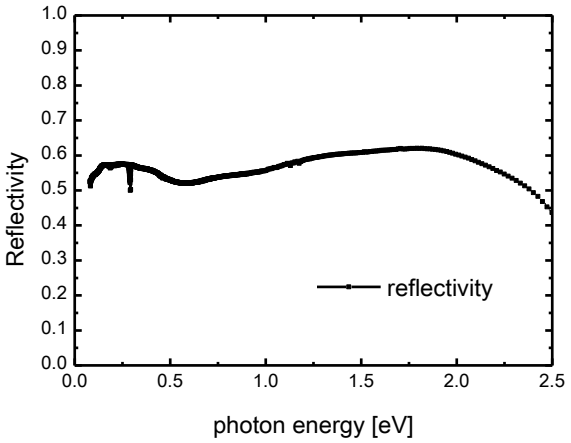

FIG. S3. IR to visible reflectivity spectrum at room temperature. The reflectivity spectrum was measured between 0.07 and 2.5 eV.

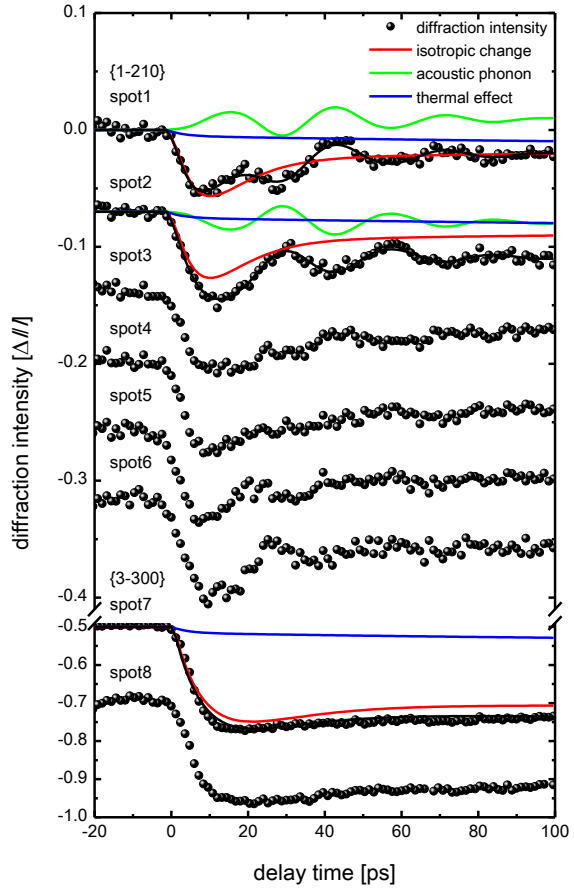

FIG. S4. The time evolution of the electron diffraction intensity from $\{1\bar{2}10\}$ planes corresponds to spot 1–6 and those from $\{3\bar{3}00\}$ planes corresponds to spot 7 & 8. The blue, green and red solid lines shown for spot 1, 2 & 7 indicate the contributions of Debye-Waller thermal effect, acoustic phonons and symmetric atomic displacement, respectively.

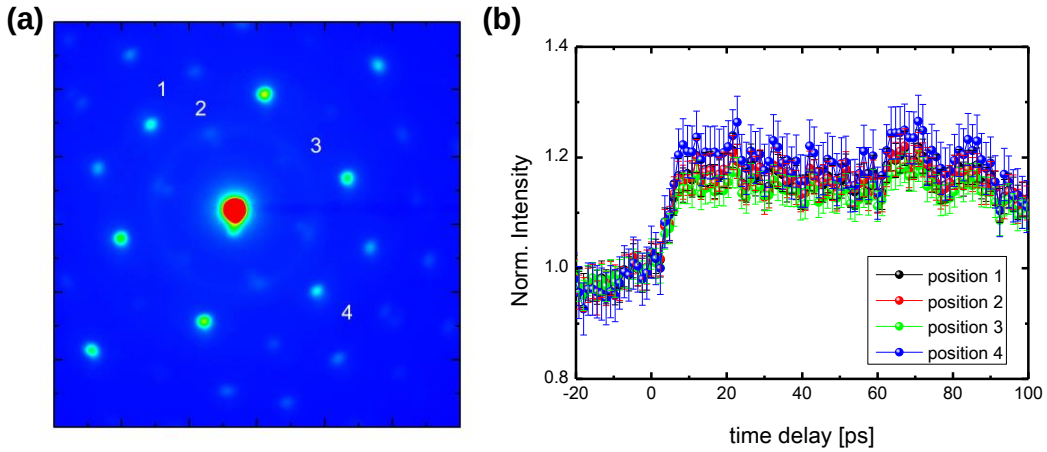

FIG. S5. For the analysis of the thermal diffuse scattering, we used random points numbered 1–4 as shown in the diffraction pattern (a). (b)The time-resolved intensity changes in diffuse scattering background. The intensity increases by 20–30% after the photoexcitation. The intensity ($I_{\mathrm{diffuse}}(\boldsymbol{k})$) of first-order thermal diffuse scattering is expressed as the function of temperature (T):

$$I_{\mathrm{diffuse}}(\boldsymbol{k}) = \mathrm{const.} \times \sum_{j=1}^{N_{\mathrm{mode}}} \frac{|F(\boldsymbol{k})|^2}{\omega_j} \coth\left(\frac{\hbar\omega_j}{2k_{\mathrm{B}}T}\right),$$

where, $F(\boldsymbol{k})$, k_{B}, ω_j are structural factor, Boltzmann constant, and phonon mode which is concerned in diffuse scattering. The phonon modes in the Bi_2Te_3 are constant since it does not undergo structural phase transition in this temperature range. The intensity increase of the diffuse scattering background by 20–30% corresponds to the temperature increase of 70–100 K from this equation. This value agrees with that derived from the thermal expansion.

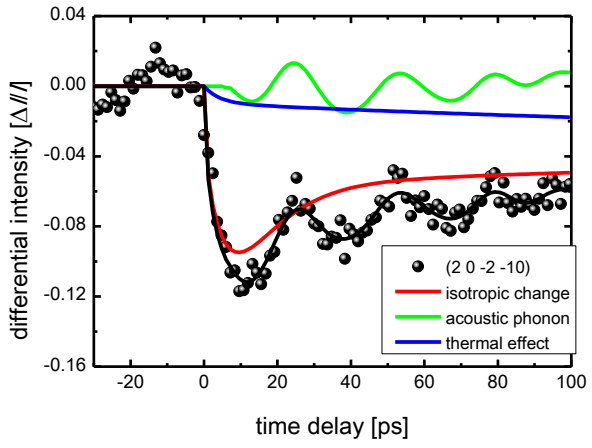

FIG. S6. The time-resolved electron intensity from $(20\bar{2}\bar{1}0)$ diffraction plane which has the *c*-axis components. The intensity shown in Fig. S6 has same signature as that from $\{1\bar{2}10\}$ planes in the main text.

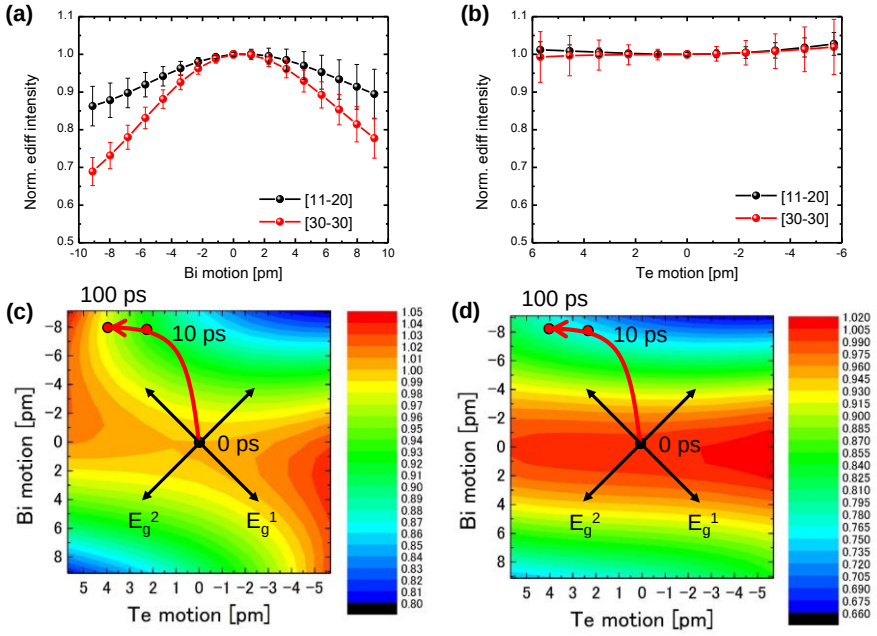

FIG. S7. Atomic displacements derived from the time-resolved electron diffraction. (a) The simulated electron diffraction intensity from $\{1\bar{2}10\}$ and $\{3\bar{3}00\}$ diffraction spots when Bi atoms symmetrically move toward the center of the unit cell along the *ab*-plane with fixing Te^1 atoms and (b) Te^1 atoms symmetrically move toward the center of the unit cell on the ab-plane with fixing Bi atoms. The two dimensional maps of the calculated differential electron diffraction intensities from (c) $\{1\bar{2}10\}$ planes and (d) $\{3\bar{3}00\}$ planes with the dynamical diffraction theory as both Bi and Te^1 atoms move independently. The red arrow is the motion of Bi and Te^1 obtained from the time-resolved electron diffraction experiments. The black arrows indicate the direction of motions of E_g^1 and E_g^2 phonon modes, respectively.

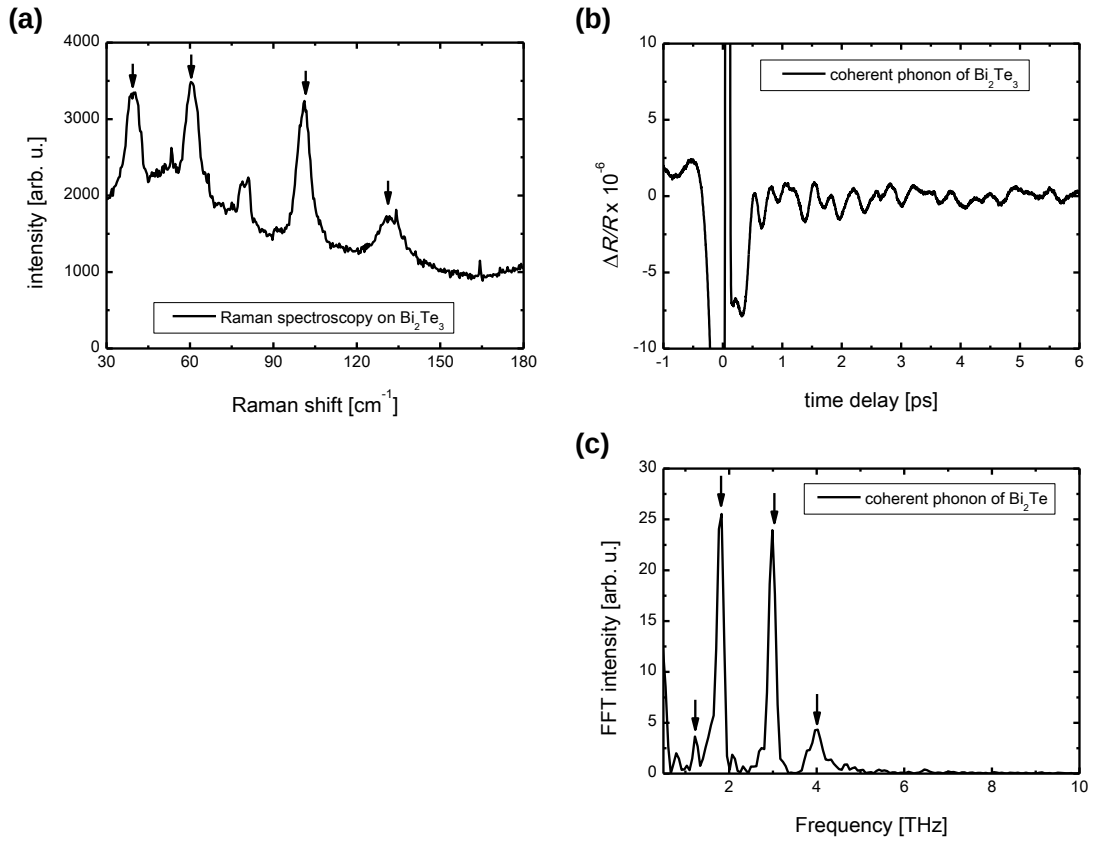

FIG. S8. The Raman spectrum from Bi_2Te_3 crystal (a) and the coherent phonon vibrations by the conventional near-IR pump-probe experiment (b). As shown in Fig S4 (a), four Raman active modes, E_g^1 (39 cm^{-1}), A_{1g}^1 (61 cm^{-1}), E_g^2 (101 cm^{-1}) and A_{1g}^2 (132 cm^{-1}) are indicated with the insert arrows. The Figure S7 (c) presents the fast Fourier transform of Fig. S7 (b), which also shows the four Raman active phonon modes (E_g^1, A_{1g}^1, E_g^2 and A_{1g}^2 modes).

Table S1. Undisturbed atomic coordinates of Bi_2Te_3 crystal resided as the space group R3m. Lattice parameters: a = 4.3835 Å, c = 30.4870 Å.

	atom	x	y	z
1	Te	0	0	0
2	Bi	0	0	0.399
3	Te	0	0	0.792

Table S2. Equivalent undisturbed atomic coordinates of Bi_2Te_3 crystal in space group P1. Lattice parameters: $a = b$ = 4.3835 Å, c = 30.4870 Å, $\alpha = \beta = 90^\circ$, and $\gamma = 120^\circ$.

	atom	x	y	z
1	Te	0	0	0
2	Bi	0	0	0.399
3	Te	0	0	0.792
4	Te	0.6667	0.3333	0.3333
5	Te	0.3333	0.6667	0.6667
6	Bi	0.6667	0.3333	0.7323
7	Bi	0.3333	0.6667	0.0657
8	Te	0.6667	0.3333	0.1253
9	Te	0.3333	0.6667	0.4587
10	Bi	0	0	0.601
11	Te	0	0	0.208
12	Bi	0.3333	0.6667	0.2677
13	Bi	0.6667	0.3333	0.9343
14	Te	0.3333	0.6667	0.8747
15	Te	0.6667	0.3333	0.5413

Table S3. Disturbed atomic coordinates of Bi_2Te_3 crystal at 10 ps as the space group of P1. Lattice parameters are same as Table II.

	atom	x	y	z
1	Te	0	0	0.0001
2	Bi	-0.007	-0.014	0.399
3	Te	0.004	0.002	0.792
4	Te	0.6667	0.3333	0.3332
5	Te	0.3333	0.6667	0.6666
6	Bi	0.6807	0.3403	0.7323
7	Bi	0.3263	0.6737	0.0657
8	Te	0.6647	0.3353	0.1253
9	Te	0.3313	0.6627	0.4587
10	Bi	-0.014	-0.007	0.601
11	Te	0.002	0.004	0.208
12	Bi	0.3403	0.6807	0.2677
13	Bi	0.6737	0.3263	0.9343
14	Te	0.3353	0.6647	0.8747
15	Te	0.6627	0.3313	0.5413

Table S4. Disturbed atomic coordinates of Bi_2Te_3 crystal at 100 ps as the space group of P1. Lattice parameters are same as Table II.

	atom	x	y	z
1	Te	0	0	0.0001
2	Bi	-0.007	-0.014	0.399
3	Te	0.007	0.0035	0.792
4	Te	0.6667	0.3333	0.3332
5	Te	0.3333	0.6667	0.6666
6	Bi	0.6807	0.3403	0.7323
7	Bi	0.3263	0.6737	0.0657
8	Te	0.6632	0.3368	0.1253
9	Te	0.3298	0.6597	0.4587
10	Bi	-0.014	-0.007	0.601
11	Te	0.0035	0.007	0.208
12	Bi	0.3403	0.6807	0.2677
13	Bi	0.6737	0.3263	0.9343
14	Te	0.3368	0.6632	0.8747
15	Te	0.6597	0.3298	0.5413

Table S5. Equivalent undisturbed atomic coordinates of Bi_2Te_3 crystal in space group of B1. Lattice parameters: a = 7.58640 Å, b = 30.47997 Å, c = 4.38000 Å.

	atom	x	y	z
1	Bi	0.1667	0.2677	0.5
2	Bi	0.1667	0.0657	0.5
3	Bi	0	0.601	0
4	Te	0	0	0
5	Te	0.1667	0.6666	0.5
6	Te	0.1667	0.4587	0.5
7	Te	0.1667	0.8747	0.5
8	Te	0	0.792	0

Table S6. Disturbed atomic coordinates of Bi_2Te_3 crystal at 10 ps as the space group of B1. Lattice parameters are same as Table V.

	atom	x	y	z
1	Bi	0.17018	0.2677	0.51058
2	Bi	0.17368	0.0657	0.50008
3	Bi	0.0035	0.601	0.9895
4	Te	0	0	0
5	Te	0.16667	0.6666	0.5
6	Te	0.16568	0.4587	0.49708
7	Te	0.16467	0.8747	0.50008
8	Te	0.999	0.792	0.003

Table S7. Disturbed atomic coordinates of Bi_2Te_3 crystal at 100 ps as the space group of B1. Lattice parameters are same as Table V.

	atom	x	y	z
1	Bi	0.17018	0.2677	0.51058
2	Bi	0.17368	0.0657	0.50008
3	Bi	0.0035	0.601	0.9895
4	Te	0	0	0
5	Te	0.16667	0.6666	0.5
6	Te	0.16493	0.4587	0.49483
7	Te	0.16318	0.8747	0.50008
8	Te	0.99825	0.792	0.00525