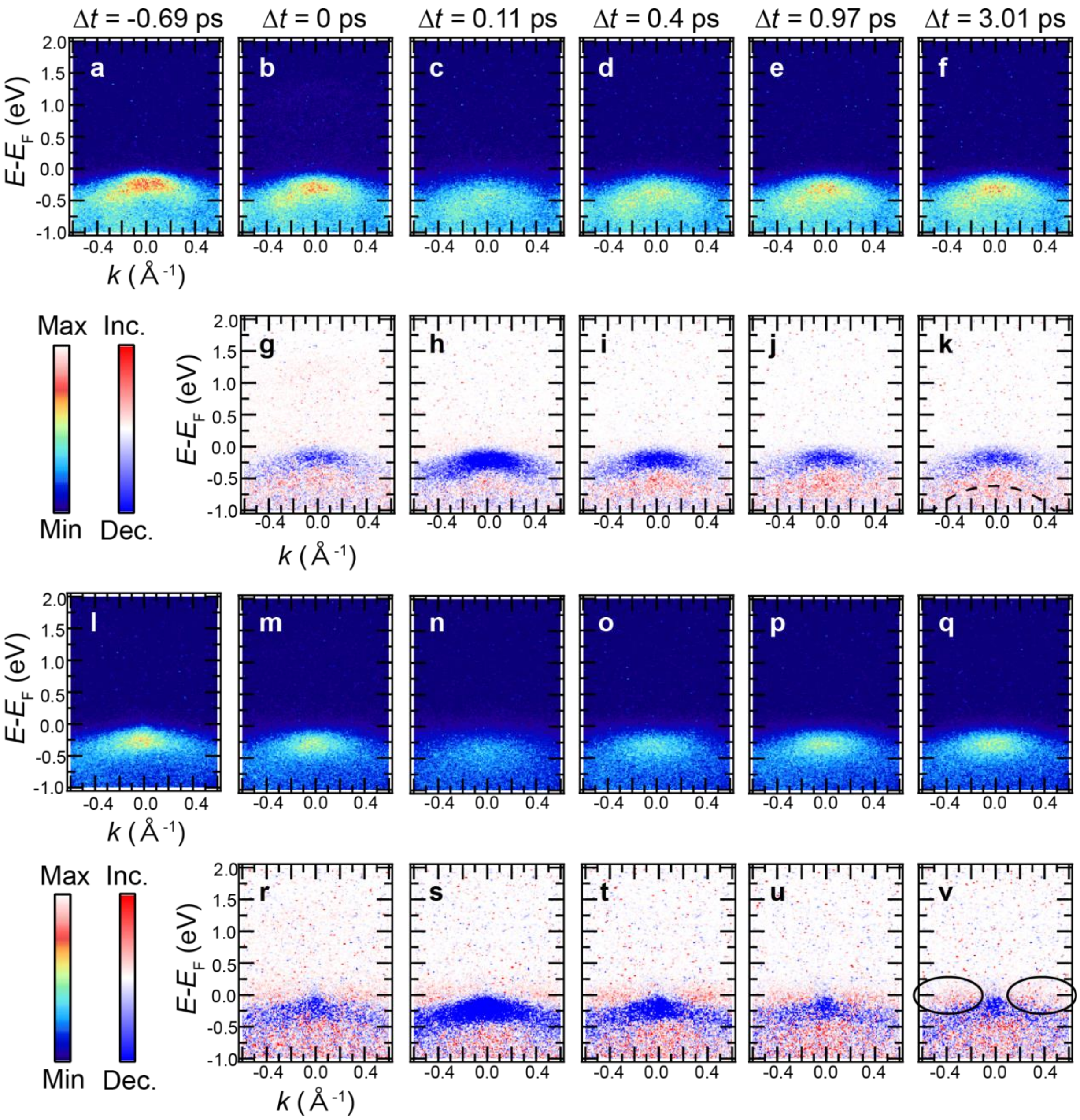
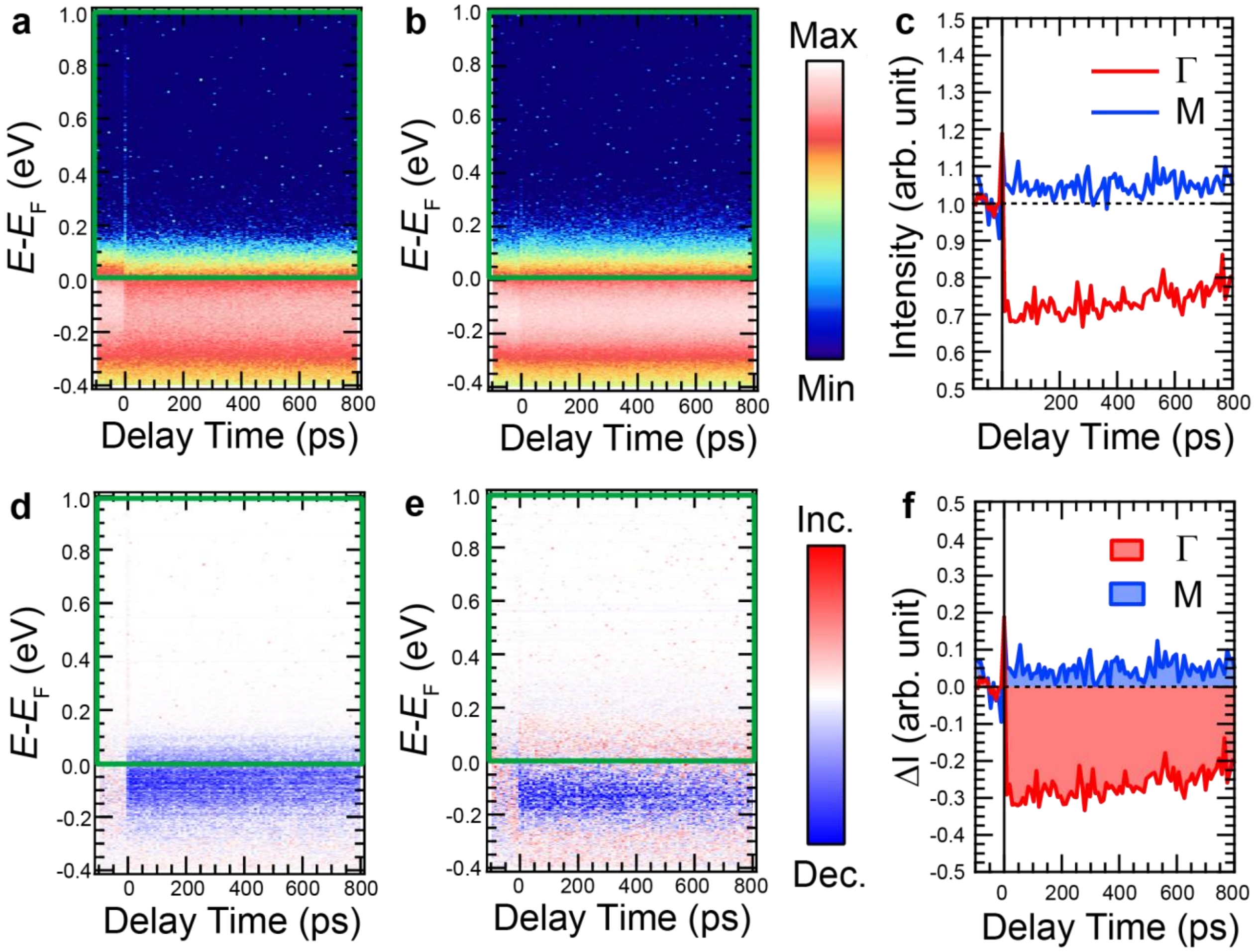
Supplementary Information

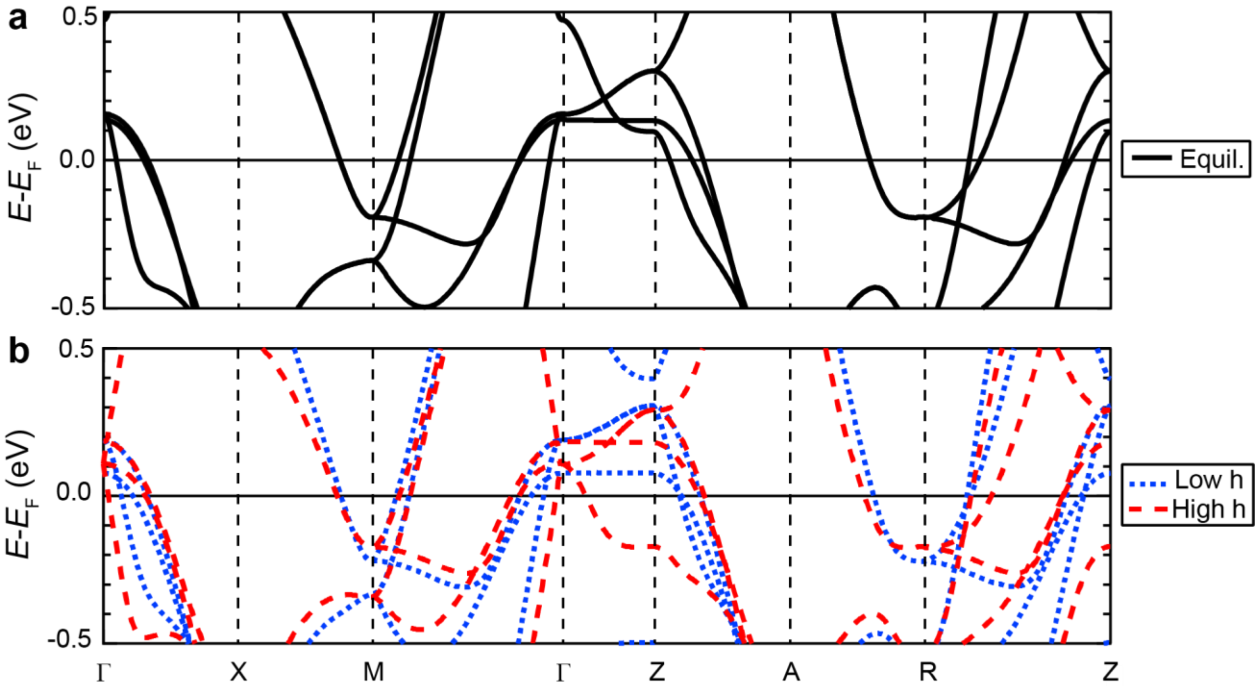
Supplementary Figures



Supplementary Figure 1 Time- and angle-resolved photoemission spectra for Γ (**a-f**) and M (**l-q**) points at selected pump-probe delays (Δt). Difference spectra for Γ (**g-k**) and M (**l-q**) points at the corresponding delay times given in (**b-f**) and (**r-v**) obtained by subtracting (**a**) and (**r**) from each spectrum, respectively. The black dashed line in Supplementary Fig. 1k indicates the band downward shift at the Γ point while the black solid circles in Supplementary Fig. 1v show the increase of electrons in electron pockets at the M point.



Supplementary Figure 2 **a**,**b**, Time-resolved photoemission spectra for a long-delay time measurement taken at around the Γ and M points, respectively. The colors are shown in log scales. **c,** Integrated photoemission intensities above E_F corresponding to the surrounded regions by green boxes in **a** and **b**, respectively. **d**,**e**, Difference spectra for Γ and M points, respectively, obtained by subtracting spectra before arrival of pump. **f**, Integrated photoemission intensities above E_F corresponding to the surrounded regions by green boxes in **d** and **e**, respectively.



Supplementary Figure 3 Calculated band dispersions for (**a**) the equilibrium structure and (**b**) the modulated structure with ±5% variation of the Se height.

Supplementary Discussion

Superconducting Gaps Extracted from LEM Shifts

Taking into account the carrier conservation, the decrease and increase of electron populations in the hole (ΔN_{h}) and electron pockets (ΔN_{e}) are the same amount after electrons are transferred from the hole to electron pockets. They are proportional to the effective mass of electrons due to the two dimensional density of states. Given the chemical potential shifts denoted as $\Delta\mu_{\mathrm{h}}$ and $\Delta\mu_{\mathrm{e}}$, ΔN_{h} and ΔN_{e} are

$$|\Delta N_{\mathrm{h}}| = |\Delta N_{\mathrm{e}}|, \tag{1}$$

$$\Delta N_{\mathrm{h}} = D_h(\mu)\Delta\mu_{\mathrm{h}}, \tag{2}$$

$$\Delta N_{\mathrm{e}} = D_e(\mu)\Delta\mu_{\mathrm{e}}, \tag{3}$$

where $D_{h/e}(\mu)$ stands for the density of states of the hole/electron band, and $D_{h/e}(\mu) \propto m_{h/e}$, where $m_{h/e}$ is the effective mass of the hole/electron band. Furthermore, by considering superconducting gaps as well, additional shifts corresponding to the averaged superconducting gap $\langle\Delta\rangle$, LEM shifts for the hole and electron pockets, LEM_{h} and LEM_{e} are

$$LEM_{\mathrm{h}} = \Delta\mu_{\mathrm{h}} - \langle\Delta\rangle, \tag{4}$$

$$LEM_{\mathrm{e}} = \Delta\mu_{\mathrm{e}} - \langle\Delta\rangle. \tag{5}$$

From Eqs (1)-(5), $\langle\Delta\rangle$ is expressed by the LEM shifts as

$$\langle\Delta\rangle = -\frac{m_{\mathrm{h}}LEM_{\mathrm{h}} + m_{\mathrm{e}}LEM_{\mathrm{e}}}{m_{\mathrm{h}} + m_{\mathrm{e}}}. \tag{6}$$