

Supplementary Materials for

Band structure evolution during the ultrafast ferromagnetic-paramagnetic phase transition in cobalt

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section S1. Time- and angle-resolved photoemission spectroscopy. Prior to our time- and spin-resolved photoelectron spectroscopy experiments, we performed time- and angle-resolved photoelectron spectroscopy for our conditions. In these measurements, we did see that the pump-excitation is homogenous throughout the accessible k_{\parallel} range ($k_{\parallel} < \pm 0.27 \text{ \AA}^{-1}$), and also that the subsequent dynamics in $\bar{\Gamma}$ - \bar{X} direction is momentum-independent. This is expected and can be explained by the small dispersion of the majority $\Delta_{2,\text{up}}$ -band and the minority $\Delta_{5,\text{down}}$ -band in this $E(k)$ region (see Fig. 2C in manuscript and Ref. [32]). Therefore, in our spin-resolved photoelectron spectroscopy experiments, we integrated over $k_{\parallel} = \pm 0.27 \text{ \AA}^{-1}$. Figure S1 shows the according angle-resolved data before excitation as well as an energy distribution curve (EDC) at the $\bar{\Gamma}$ -point

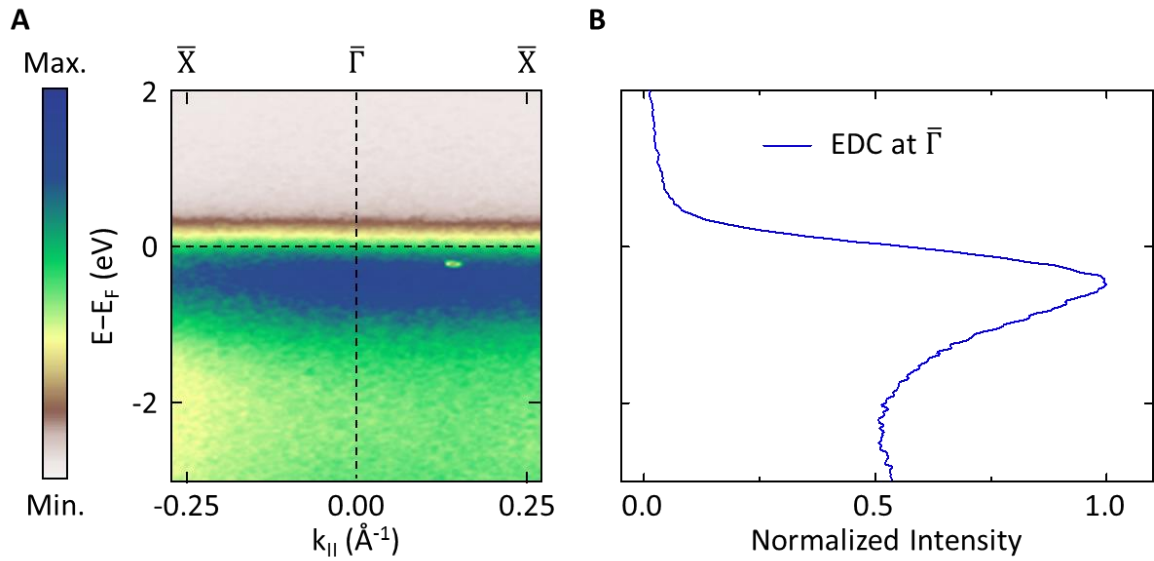


fig. S1. Time- and angle-resolved photoemission spectroscopy. (A) ARPES intensity map of 30 ML of Co on a Cu(001) substrate. Close to the Fermi-level we see the contributions of the Co 3d bands and a surface resonance (blue intensity area) along the complete momentum range. (B) EDC of the same ARPES map extracted at the $\bar{\Gamma}$ -point, marked by the vertical dashed line.

section S2. Time- and angle-resolved photoemission spectroscopy: Difference spectra.

Figure S2(A) shows a difference spectrum for a spectrum taken at -200 fs before excitation and a spectrum taken at 50 fs after excitation. In addition, fig. S2(B) displays the corresponding difference EDCs around the $\bar{\Gamma}$ -point as a function of delay time. These difference spectra clearly illustrate the homogenous excitation in momentum space, and also the momentum-independent dynamics. Furthermore, this measurement illustrates that the pump-excitation affects states for a range of ± 0.5 eV around the Fermi-level. To exclude the direct effect of the pump excitation in the band-mirroring analysis, the fitting routine was carried out for states with binding energies >0.5 eV.

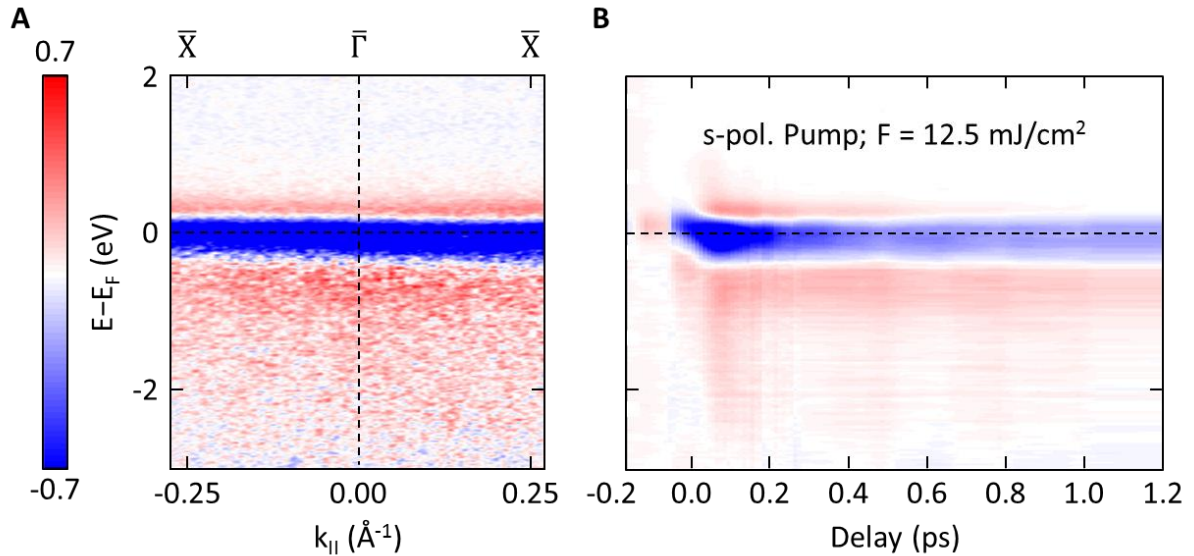


fig. S2. Time- and angle-resolved photoemission spectroscopy: Difference spectra. (A) ARPES difference map of spectra taken at 200 fs before and 50 fs after the arrival of the pump pulse. Red areas denote an increase in photoelectron intensity while blue areas denote a decrease. (B) Momentum integrated intensity difference as a function of $E-E_F$ and time delay of pump and probe pulses.