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## Femtosecond spin-dependent charge transfer at Co/Cu(001) interfaces

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Controlling ultrafast charge and spin dynamics in solids with ultrashort laser pulses requires us to first identify the quantum processes relevant to light-matter interaction on femtosecond timescales. We disentangle the elementary processes behind ultrafast spin transfer at epitaxial Co/Cu(001) interfaces due to optical excitation with 1.5 eV photon energy by combining femtosecond time-resolved interface-sensitive magnetization-induced second harmonic generation and ab initio time-dependent density functional theory. Finding a convincing agreement between the observables in theory and experiment, we directly identify spin-dependent charge transfer between Co and Cu active at  $< 30$  fs, and spin-flips mediated by the spin-orbit interaction, which lead to a loss of spatially integrated spin polarization and dominate at  $> 30$  fs.

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