Tunable Competing Optical Excitation Pathways in the Topological Surface States of Bi_2Te_3

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Topological insulators (TIs) have topological surface states (TSSs) that exhibit Dirac-cone-like dispersion and helical spin textures in momentum space [1]. The ultrafast optical spin response of such TSSs has attracted considerable attention in optospintronics, which aims at optical control of spin-polarized electrons. While various optical responses of TSSs have been reported, elucidating their ultrafast mechanisms requires an understanding of optical coupling involving unoccupied intermediate states. Pump-probe time- and angle-resolved photoemission spectroscopy (tr-ARPES) has been widely employed to study nonequilibrium population dynamics of TSSs in energy and momentum space [2]. By contrast, owing to its intrinsically coherent multiphoton excitation, angle-resolved two-photon photoemission spectroscopy (2PPE-ARPES) directly probes transient optical coupling via unoccupied intermediate states and the associated dephasing dynamics within an ultrashort pulse. Under resonant conditions, competing excitation pathways can produce characteristic spectral modulations and even transient band renormalization [3]. However, experimental studies of coherent optical phenomena in TSSs have remained limited, with only a few reports so far [4, 5].

In this work, we introduced a femtosecond ultrashort-pulse laser into the spin- and angle-resolved photoemission spectroscopy (Spin-ARPES) system developed at the Hiroshima Synchrotron Radiation Center (HiSOR) [6] and performed 2PPE-ARPES measurements on the TSSs of the topological insulator Bi_2Te_3. Using 2PPE-ARPES, we identify two distinct optical excitation pathways in the TSSs of Bi_2Te_3: an off-resonant transition mediated by virtual states [Fig. 1(a)] and a resonant transition via real unoccupied intermediate states [Fig. 1(b)]. Coherent competition between excitation pathways manifests as an intensity modulation featuring a distinct node at a specific kinetic energy in the 2PPE-ARPES spectra [green arrows in Fig. 1(c)]. Photon-energy-dependent measurements reveal that the spectral feature above the node reflects dispersions of unoccupied intermediate states, while that below the node follows those of the occupied TSS [Fig. 2]. This supports an interpretation in terms of competing resonant and off-resonant excitation pathways. Furthermore, temperature-dependent measurements show that a shift of the chemical potential with increasing temperature modifies the intensity of the resonant channel [Fig. 3(a, b)]. As a result, temperature provides a knob for selective on/off control of the resonant excitation pathway [Fig. 3(d, e)]. These results provide microscopic insight into the optical excitation mechanisms of TSSs and highlight the potential for controlling their optical responses, relevant for future spintronic devices.

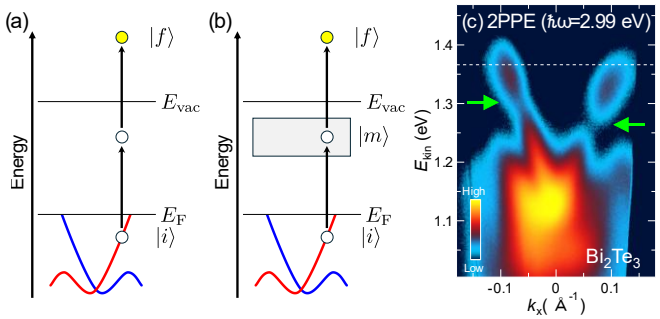

FIGURE 1. (a, b) Schematic illustration of the two competing paths of single-color 2PPE processes: (a) off-resonant transition from the occupied TSS ($|i\rangle$) to the photoelectron final state ($|f\rangle$) via a virtual state, and (b) resonant transition through unoccupied intermediate states ($|m\rangle$). (c) 2PPE-ARPES results at T= 20K. The green arrow marks an intensity node where the 2PPE signal is suppressed.

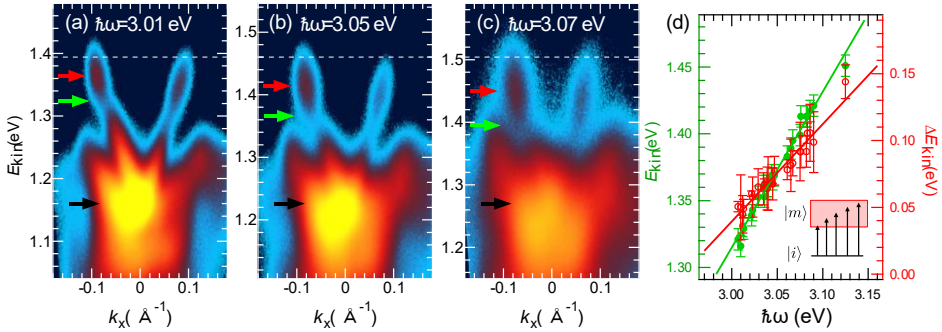

FIGURE 2. (a)-(c) Representative 2PPE-ARPES results acquired at T= 20K with various excitation $\hbar\omega$. The energies at which the intermediate-state bands, the intensity node, and the Dirac point (DP) appear are indicated by red, green, and black arrows, respectively. (d) Plot of the E_{kin} position of the intensity node (green) as a function of $\hbar\omega$, together with the observable energy window of the intermediate-state feature in the 2PPE spectra (red). The solid lines are linear fits to the experimental data, yielding slopes of 1.1±0.1 (green) and 0.7±0.1 (red).

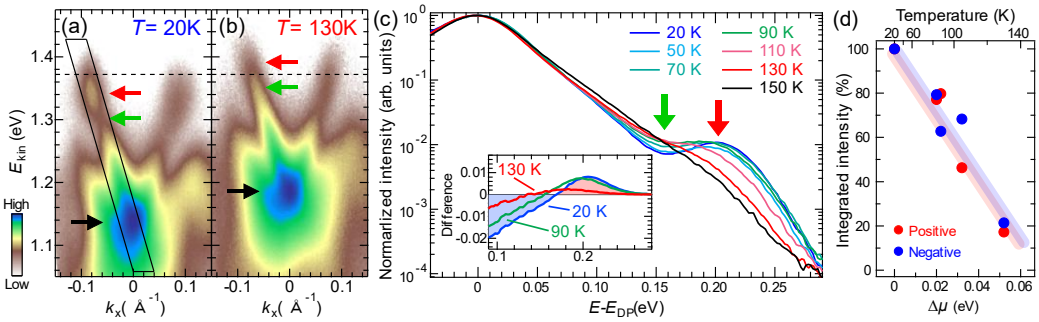

FIGURE 3. (a, b) The representative 2PPE-ARPES results with $\hbar\omega$= 3.00 eV at 20K (a) and 130K (b). The energies at which the intermediate-state bands, the intensity node, and DP appear are indicated by red, green, and black arrows, respectively. (c) The logarithmic plots of the integrated 2PPE signals along the TSS dispersion within the E-k window denoted by the parallelogram in (a). To compensate for the temperature-induced shift of the chemical potential, the horizontal axis is referenced to the DP energy (E-E_{DP}). The red and green arrows indicate the energy positions of the peak and dip intensities. The inset shows the differential intensities with respect to the data at T= 150K where the 2PPE signals of the intermediate state are negligibly small. (d) Normalized area intensities of the positive and negative components extracted from the difference spectra [inset of (c)]. The areas are obtained by integrating both parts and are normalized to their respective values at 20K. The horizontal axis represents the relative shift of the chemical potential with respect to its position at 20K, $\Delta\mu$, estimated from the T-induced shift of E_{DP}. The shaded lines are guides to the eye.

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