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Circularly polarized light interaction in topological insulators investigated by time-resolved ARPES

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Abstract. Topological Insulators (TI) represent a hot-topic for both basic physics and promising applications because of the in-plane spin-polarized surface states (TSS) arising within the bulk insulating energy gap. The backscattering protection and the control of the spin polarization using ultrashort light pulses open new scenarios in the use of this class of materials for future opto-spintronic devices. Using time- and angle-resolved photoemission spectroscopy on $Sb_xBi_{(2-x)}Se_yTe_{(3-y)}$ class we studied the response of spin-polarized electrons to ultrashort circularly-polarized pulses. Here, we report for the first time the experimental evidence of a direct coupling between light and empty topological surface states (ESS) and the establishment of a flow of spin-polarized electrons in **k**-space i.e. a photon-induced spin-current.

1. Introduction

In the last years, topological insulators have become a hot-topic as benchmark for novel theoretical and experimental studies in spintronics. However, the electronic dynamics induced by ultrashort laser pulses is far from being completely unravelled in these materials. Because circularly-polarized light couples to the total angular momentum of electron (i.e. orbital momentum + spin), we performed time and angle-resolved photoemission spectroscopy (TR-ARPES) measurements using linear p-polarized pump beam and circularly-polarized probe beam. In this way we can reveal a spin-related parameter of the spin-polarized band structure. In Sec. 3.1 we present our circular dichroism (CD) TR-ARPES measurements on the topological insulator Bi_2Te_3. In Sec. 3.2 we show our measurements with circularly-polarized pump and linear p-polarized probe on the Bi_2Se_3 topological insulator. In this configuration the selective-coupling of helicities of light with spin leads to a spin-selective pumping.

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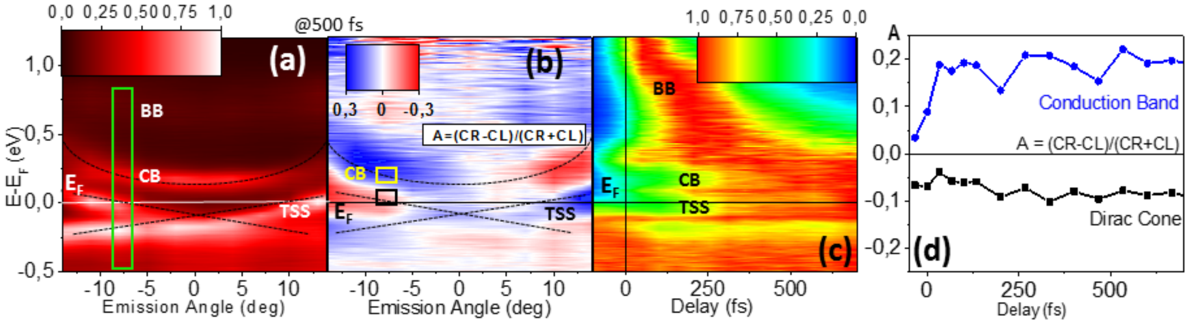

Figure 1. (a) Bi_2Te_3 ARPES map binding energy vs. angle of emission at 500 fs pump-probe delay. (b) Dichroic asymmetry $A = (CR - CL)/(CR + CL)$ map binding energy vs. angle of emission computed for (a). The colored squares represent the angles of emission and the energies at which the temporal evolutions of A shown in (d) are extracted. (c) Log intensity map binding energy vs pump-probe delay at the angle of emission marked by the green rectangle in (a). E_F:Fermi energy. Dashed black lines are a guide for eyes to follow linear dispersion of the Dirac cone and the parabolic dispersion of the conduction band.

2. Materials and methods

Our Yb-based TR-ARPES setup, described in detail in Ref. [1], provides pump pulses shorter than 30 fs at 1.85 eV and probe pulses at 6.02 eV with 65 fs time duration by means of a cascade of non-linear processes. The temporal resolution is ≈70 fs. Pump and probe beams impinge almost collinearly on the sample at an angle of about 45°. Photoemitted electrons are detected by means of a time-of-flight analyzer with energy and angular resolutions of ≈ 50 meV and 0.8°, respectively. High quality single crystals TIs were grown by the self-flux method. Powders of bismuth, selenium and tellurium were stoichiometrically mixed, put in alumina crucibles and sealed in a quartz tube under vacuum. The materials were heated to 1000° C for 12 hours and slowly cooled down to 500° C over 100 hours before reaching room temperature [2]. The resulting crystals, belonging to the $R\bar{3}m$ space group, were cleaved in situ at pressure <5 x 10^{-10} mbar for the TR-ARPES measurements. They naturally cleave along the (111) plane. All measurements here reported were performed at room temperature. Pump fluence was ≈ 0.5 $\frac{mJ}{cm^2}$.

3. Results

3.1. Circular Dichroism of probe on Bi_2Te_3

Fig. 1(a) shows ARPES map measured on Bi_2Te_3 for one helicity of the probe (circular right (CR)) and at 500 fs pump-probe delays. We see the parabolic-dispersing conduction band (CB) and the linear-dispersing topological surface state (TSS). In addition, a less-intense bulk band (BB) above the Fermi level at 0.8 eV binding energy can be observed. The binding energy vs. pump-probe delay map (Fig. 1(c)) for one emission angle (green rectangle in Fig. 1(a)) highlights the pump role and the dynamics of the electrons. The pump promotes electrons into high-energy bulk bands (BB) and after 100 fs they decay filling the lower energy empty bands i.e. the CB and the TSS. As already reported [3, 4], TSS is not directly populated by the pump beam. Computing the dichroic asymmetry $A = (CR - CL)/(CR + CL)$, we have a signature of the spin-polarization of the bands. As shown in Fig. 1(b), the two branches of the TSS present opposite dichroic signals passing throught the Γ-point. The asymmetry map also exhibits a dichroic contrast in the CB, always opposite to the signal in the upper part of the cone. We found a common trend of the dichroism in several compounds of $Sb_xBi_{(2-x)}Se_yTe_{(3-y)}$ class (not

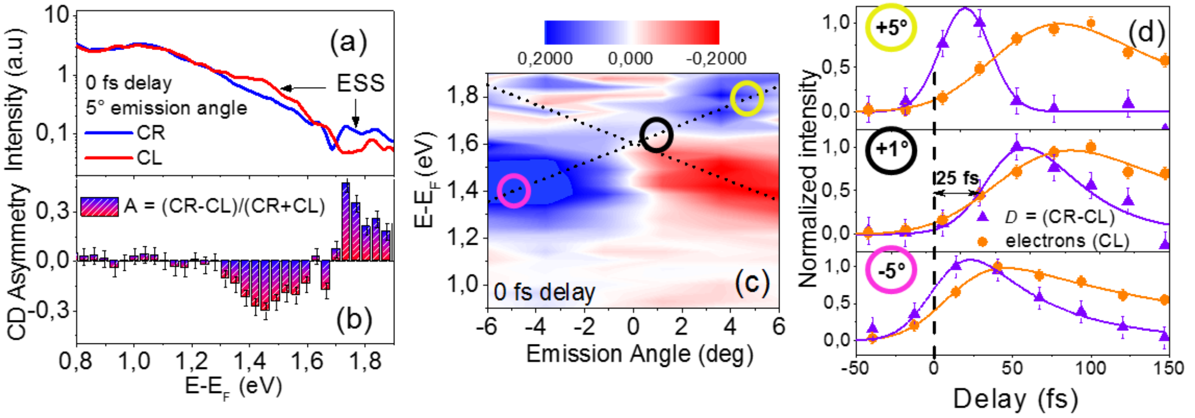

Figure 2. (a) Bi_2Se_3 energy distibution curves in the 0.8-1.9 eV binding energy range for two opposite pump helicities (circular right CR (blue) and circular left CL (red)) at 0 fs delay. (b) Dichroic asymmetry computed for (a). Dashed black lines are a guide for eyes to follow linear dispersion of the empty surface state (ESS). (c) Dichroic asymmetry map binding energy vs. angle of emission at 0 fs. The colored circles represent the angles of emission and the energies at which the CD difference and the related electronic background shown in (d) are calculated. (d) Comparison between dichroism (violet triangles) and the unpolarized electronic background induced by CL pump (orange circles) at three different angles of emission +5° (top), +1° (middle) and 5°(bottom) along the CR-pumped branch (blue branch in (c)).

shown) suggesting a universal property. A spin-polarized signature in the CB has already been reported as a surface-resonant state in Bi_2Se_3 [5] and in Sb_2Se_3 [6]. The dynamics of the dichroic signal of the two bands (Fig. 1(e)) suggests that the signal arising from TSS and from CB have different physical origin. For a spin-polarized band the dichroic asymmetry is expected to be constant in time. The dichroic signal of the TSS is constant at all delays confirming that it is a truly spin-polarized band. In contrast, the temporal evolution of the CB dichroism presents a rise around zero that resembles the relaxation-assisted population of the band. The electrons promoted in BB are unpolarized: when they decay, only the electrons matching the spin of the empty branch of TSS can fill it. The electrons of the opposite spin do not accumulate in BB due to the high energy and they decay into the first lower energy accessible band, i.e. the conduction band. Our data suggest the presence of a spin-dependent decay channel that leads to a polarization of the conduction band.

3.2. *Selective circularly-polarized pumping of Bi_2Se_3*

In TR-ARPES the main effect of the pump beam is to promote electrons into unoccupied bands by optical transitions. If the pump beam is circularly-polarized, it has been showed that this process becomes spin-selective [7]. In Fig. 2(a) the electron distribution curves (EDCs) of intrinsically n-doped Bi_2Se_3 for two different pump-helicities are reported. The unpolarized bulk bands are equally filled by the two opposite pump-helicities while, in the high-energy decay tails of these bands, two shoulders appear. Looking at the circular-asymmetry in Fig. 2(b), the two shoulders exhibit opposite character. In addition, they reverse their sign passing through the Γ-point (Fig. 2(c)) and a Dirac cone-like linear dispersion appears. This observation is consistent with previously reported 2PPE-measurements that disclosed an empty topological surface state (ESS) at the same binding energy [8, 9]. Focusing on the most intense band (blue one in Fig. 2(c)), we studied the dynamics of the spin-selectively promoted electrons by the circular-difference $D = C_R - C_L$. As shown in Fig. 2(a), only one helicity is able to promote electrons in the ESS while both are populating the underlying bulk bands. The circular-difference D then represents the spin-ordered electronic population promoted in ESS at the net of the bulk one. The dynamics for D and for the electrons of the bulks for three different angles (marked by different colored circles in Fig. 2(c)) are reported in Fig. 2(d). We see that for outer angles ($\pm 5°$), i.e. larger $\mathbf{k}_{||}$, the dynamics of the electrons promoted in ESS starts 25 fs earlier with respect to the region close to the Γ -point (i.e. 1°). This implies that the circular-pump is promoting a direct transition into the ESS only at outer angles starting from electrons occupying the valence and conduction bands. The spin-polarized electrons promoted into the ESS flow by electron-electron scattering along the linear branch of the Dirac cone without any spin-order dissipation and then, after 25 fs, the dichroic signal arises in the region close to the Γ-point. Our results highlight the capability to directly excite electrons in the ESS by circular light and, in addition, they show the establishment of a flow of spin-polarized electrons in **k**-space, i.e. a spin-current.

4. Conclusions

We have presented a circular dichroism time-resolved photoemission study on the Bi_2Te_3 topological insulator. We have observed a dichroic signal in the conduction band and studying the temporal evolution of this signal, we clarified that it originates from the presence of a spin-dependent decay channel for photon-excited electrons. Our results suggest the capability to generate a spin-polarized electron accumulation in the conduction band in addition to the spin-polarized topological surface state. We have presented also spin-selective pumping by time-resolved photoemission measurements on the topological insulator Bi_2Se_3. We reported a population of the empty topological surface state (ESS). The **k**-dependent temporal evolution of the electronic population in the ESS suggests an intraband-flow of spin-polarized electrons along the Dirac cone, i.e. a photon-induced spin-current.

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