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# Probing spin chirality of photoexcited topological insulators with circular dichroism: multi-dimensional time-resolved ARPES on $\text{Bi}_2\text{Te}_2\text{Se}$ and $\text{Bi}_2\text{Se}_3$

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Using time-resolved multi-dimensional Angle-Resolved Photoelectron Spectroscopy (ARPES) we explore the angular momentum transfer of low energy polarized photons to two prototyp topological insulators,  $\text{Bi}_2\text{Te}_2\text{Se}$  and  $\text{Bi}_2\text{Se}_3$ . Our comparative study is based on the analysis of circular dichroism in the photoemission yield of photoexcited Dirac states, and reveals that the spin vector of in-gap Dirac electrons in  $\text{Bi}_2\text{Te}_2\text{Se}$  presents a more pronounced out-of-plane component compared to that of  $\text{Bi}_2\text{Se}_3$ . We show that the multi-dimensional ARPES approach can be effectively used to observe the spin texture of photoexcited topological insulators, and to unambiguously disentangle experimental geometry and matrix element effects.

## I. INTRODUCTION

Three dimensional topological insulators (TIs) are bulk insulators with conducting Dirac surface states (DSS) protected by time-reversal symmetry[1]. One of the essential features of their topological states is spin-momentum locking: as theoretically predicted [2–7] and soon experimentally confirmed [8–11] DSS possess a helical spin texture, i.e. the spin of the electrons is locked to their momentum and electrons with opposite momentum have opposite spin. All the most promising technological perspectives for TIs [12] are intimately related to this feature, because it implies that back-scattering of electrons in DSS induced by non-magnetic impurities is suppressed [13, 14]. Consequently, the measurement of spin texture in momentum space is a crucial element for the experimental study of the electronic structure of TIs.

Among the methods that give simultaneously access to both spin and wavevector, spin-resolved ARPES allows a direct measurement of the electron spin [15–17]. An alternative approach is represented by Circular Dichroism (CD) in the photoemission yield, that has also been extensively used for TIs [11, 18–22], even though extracting quantitative information on the electron spin is much more challenging: for instance, the information on its initial spin state can be altered by the polarization of the incident photon[23]; the spin texture of a TI system can also be affected by the surface itself [7, 24]; furthermore, due to its intrinsic surface sensitivity, at certain photon energies, ARPES can only probe a layer-integrated spin texture[25].

The main advantage of CD-ARPES - the possibility of

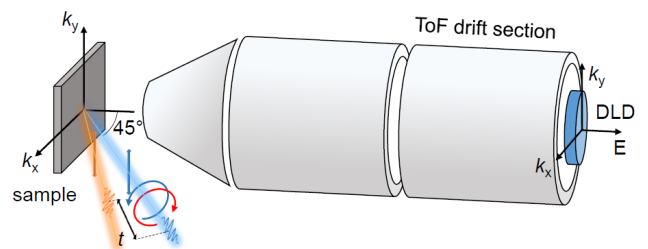


FIG. 1. Schematic of the experimental geometry

effectively providing the overall view of spin texture behavior in momentum space - can be fully exploited when performed with a multi-dimensional electron detection scheme (i.e. the simultaneous measurement of  $E$ ,  $k_x$  and  $k_y$ ). For instance, CD in the photoemission yield measured with an angle-resolved time-of-flight (ARTOF) detector has been used to probe the intrinsic spin texture of the filled electronic states of  $\text{Bi}_2\text{Se}_3$  [9]. Since with multi-dimensional detection the kinetic energy ( $E$ ) and the two components of the photoelectron momentum ( $k_x$ ,  $k_y$ ) parallel to the surface are all measured without rotating the sample, one can easily discriminate between matrix element and experimental geometry effects [9]. Extending the same approach to time-resolved ARPES makes it possible to translate all these experimental possibilities to the study of out-of-equilibrium topological materials and to their excited electronic states.

In this article, we report a comparative study of the excited electron spin texture for two prototype TIs,  $\text{Bi}_2\text{Te}_2\text{Se}$  and  $\text{Bi}_2\text{Se}_3$ , with multi-dimensional time-resolved ARPES. Our  $I(E, k_x, k_y, t)$  data provide a complete description of the strong warping characterizing the DSS above the Fermi level of  $\text{Bi}_2\text{Te}_2\text{Se}$ , in agreement

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with previous indications [26]. The analysis of the CD-ARPES evolution as a function of the electron's binding energy from the Dirac point (DP) to the bulk conduction band (BCB) indicates that the in-gap DSS of Bi<sub>2</sub>Te<sub>2</sub>Se present a strong out-of-plane spin polarization compared to those of Bi<sub>2</sub>Se<sub>3</sub> [9]. This experimental configuration makes it possible to directly observe the overall spin texture behavior of photoexcited electronic states, ruling out spurious effects related to the experimental geometry.

## II. EXPERIMENTAL

We have investigated high-quality stoichiometric Bi<sub>2</sub>Te<sub>2</sub>Se and Bi<sub>2</sub>Se<sub>3</sub> single crystals grown by the standard Bridgman-Stockbarger method using a vertical temperature gradient pull. Bi<sub>2</sub>Se<sub>3</sub> is a naturally grown n-type TI whereas the Bi<sub>2</sub>Te<sub>2</sub>Se has shown high resistivity [27] and a surface chemical potential lying within the bulk band-gap [26, 28]. Indeed, Bi<sub>2</sub>Te<sub>2</sub>Se is considered as a prototype intrinsic TI.

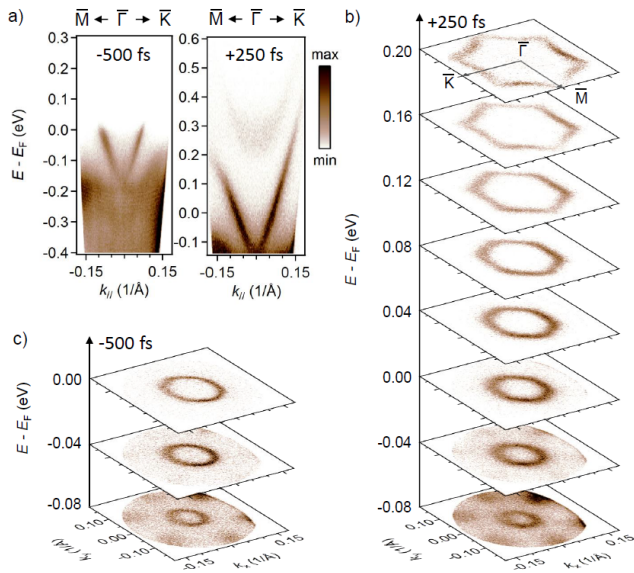


FIG. 2. (a)  $E - k$  cuts along  $\bar{\Gamma}\bar{K}$  et  $\bar{\Gamma}\bar{M}$  taken by integrating  $I(E, k_x, k_t, t)$  data in  $k$  space  $\pm 0.02\text{\AA}^{-1}$  around  $\bar{\Gamma}$  at a delay of  $t = -500$  fs and  $t = +250$  fs. (b) Isoenergy contour maps of Bi<sub>2</sub>Te<sub>2</sub>Se taken from the direct measurement of the photoelectron intensity  $I(E, k_x, k_y, t)$  with  $S$ -linear polarized light at  $t = -250$  fs and (c)  $t = -500$  fs after photo-excitation, respectively.

The multi-dimensional ARPES experiments were performed using a Scienta ARTOF 10k analyzer with an acceptance angle of  $\pm 22^\circ$ . A femtosecond Ti:Sapphire laser system FemtoSource XL 300 delivers 50 fs near-infrared pulses at 1.55 eV of photon energy and at 5 MHz of repetition rate. The repetition rate has been subsequently reduced to 500 kHz thus to make it suitable for the analyzer's Delay-Line-Detector (DLD). The near-infrared

pulses were split onto two replicas, the pump and the probe. The probe replica is used to generate ultraviolet (UV) pulses of 6.2 eV by cascade frequency mixing in  $\beta$ -BaB<sub>2</sub>O<sub>4</sub> non-linear crystals [29]. Both, the pump and probe beams were focused almost collinearly on the sample surface. The overall photoemission energy resolution is about 15 meV, the angular resolution  $0,05^\circ$ , and the time resolution about 160 fs. The measurements were conducted at a base pressure of  $2 \times 10^{-10}$  mbar. The spot-size of the UV probe beam at the sample's surface is  $\sim 40 \mu\text{m}$  while that of the pump beam is  $\sim 100 \mu\text{m}$ . The incident pump fluence was fixed at  $120 \pm 5 \mu\text{J}/\text{cm}^2$ . The samples were mounted on a cryogenic manipulator cleaved *in situ* and measured under ultra high vacuum at 85 K. At this temperature, the surface band-bending in Bi<sub>2</sub>Te<sub>2</sub>Se is below 5 meV [28] hence, below our spectral resolution.

The experimental geometry we used for CD is shown in Figure 1. Left- ( $L$ ) and right- ( $R$ ) circularly polarized photons (spiral arrows) impinge on the sample surface at an angle of at an angle  $30^\circ$ . Circularly polarized light is generated using a quarter wave-plate placed right before the last mirror that sends the UV beam into the vacuum chamber. The degree of polarization is up to  $\approx 90\%$  including birefringence from the vacuum window.

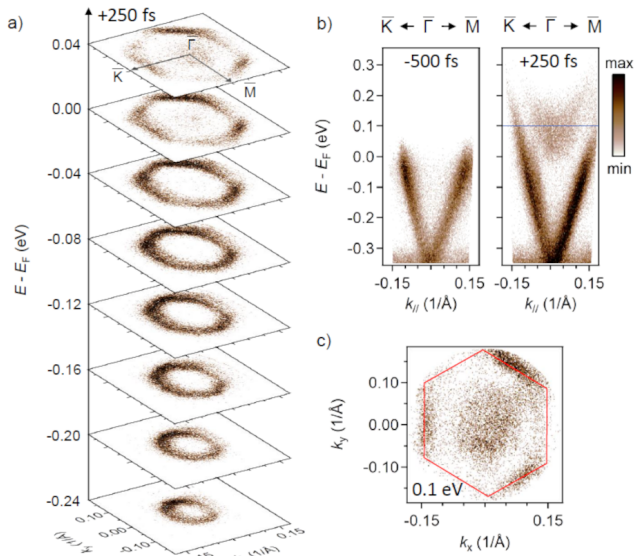


FIG. 3. (a) Isoenergy contour maps of Bi<sub>2</sub>Se<sub>3</sub> taken from the direct measurement of the photoelectron intensity  $I(E, k_x, k_y, t)$  with  $R$ -circular polarized light at  $+250$  fs after photo-excitation. (b)  $E - k$  cuts along  $\bar{\Gamma}\bar{K}$  et  $\bar{\Gamma}\bar{M}$  taken by integrating  $I(E, k_x, k_t, t)$  data in  $k$  space  $\pm 0.02\text{\AA}^{-1}$  around  $\bar{\Gamma}$  at a delay of  $t = -500$  fs and  $t = +250$  fs. (c) An isoenergy contour at  $+0.1$  eV. The red hexagon is a guide for the eye displaying the degree of warping at that energy level.

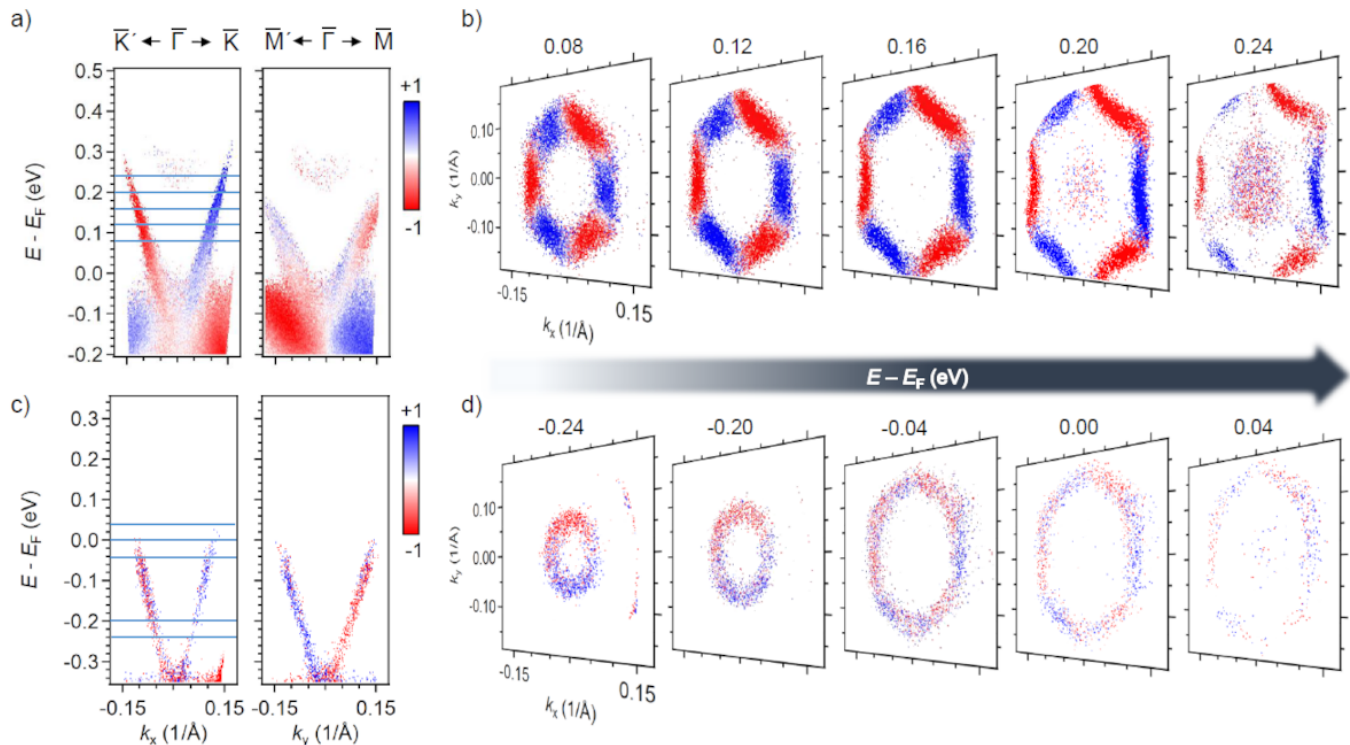


FIG. 4. (a) Normalized difference energy distribution  $\Delta I(E, k_x, k_y, t)$  maps measured using  $L$ - and  $R$ -circularly polarized light in  $\text{Bi}_2\text{Te}_2\text{Se}$  at a delay of +250 fs. (b) Difference isoenergy contour maps at different energies. (c) Similar normalized difference energy distribution maps and (d) isoenergy momentum curves in  $\text{Bi}_2\text{Se}_3$ .

### III. RESULTS AND DISCUSSION

The time-of-flight of the photo-ejected electrons from the sample surface enables the direct measurement of the photoelectron intensity as a function of energy  $E$  simultaneously over a two-dimensional momentum space that is  $I(E, k_x, k_y, t)$ . Figs. 2(b) and 2(c), respectively, show isoenergy contours from a freshly cleaved  $\text{Bi}_2\text{Te}_2\text{Se}$  surface at various energies before and after excitation with IR pulses. The chemical potential of the photo-excited surface is found at  $\approx 175$  meV (see Fig. 2(a)) above the DP, consistent with previous studies [28]. DSS show a strong degree of hexagonal warping. As a result, the isoenergy contours go from nearly circular to hexagonal for energies  $E - E_F > +0.1$  eV. For energies close to BCB the hexagon is further warped outwards forming a concave hexagon. The BCB minima along  $\bar{\Gamma}\bar{M}$  hybridize with DSS due to the threefold crystal symmetry along the [001] direction. It is worth noting that the hexagonal warping of DSS has been previously observed in Rb-covered  $\text{Bi}_2\text{Te}_2\text{Se}$  using static ARPES, exploiting the downwards band bending induced by alkali metal deposition on the surface [30]. Nevertheless, the dispersion of the two-dimensional gas on the surface follows the crystal symmetry but the subsequent potential gradient creates Rashba-type spin-split states [31]. Hence, the dispersion of the electronic states is not equivalent to that of

a pristine surface, due to a strongly anisotropic spin-orbit splitting [32]. Recently, a work on a photo-excited pristine surface [33] using time- and angular-resolved ARPES has shown isoenergy contours of unoccupied states, but without a clear indication of the aforementioned warping. To the best of our knowledge, our results provide the first clear and direct measurement of the warped DSS in  $\text{Bi}_2\text{Te}_2\text{Se}$ .

In Figure 3(a), we show isoenergy contour maps and in 3(b)  $E - k$  cut along the high symmetry directions of a freshly cleaved  $\text{Bi}_2\text{Se}_3$  surface before and after photo-excitation for comparison purposes. The chemical potential lies at  $\approx 350$  meV above the DP. Consistently with previous studies [34], DSS display circular shape for the entire energy range up to BCB minimum and it becomes warped for energies above that point. Figure 3(c) shows hexagonal warping of DSS at an energy cut of +0.1 eV recorded with  $R$ -circular polarized light. Note also that the warped DSS do not deform as much as in  $\text{Bi}_2\text{Te}_2\text{Se}$ .

It was theoretically [3, 35, 36] and experimentally shown [14] that an accentuated hexagonal warping of DSS may substantially affect the quasi-particle interference around non-magnetic impurities on the surface of TIs. Indeed, non-convex warped surfaces provide additional nesting vectors[3] enhancing Friedel oscillations around non-magnetic impurities. Detailed studies of DSS warped states will provide deeper insights on the transport properties of DSS in the presence of non-magnetic

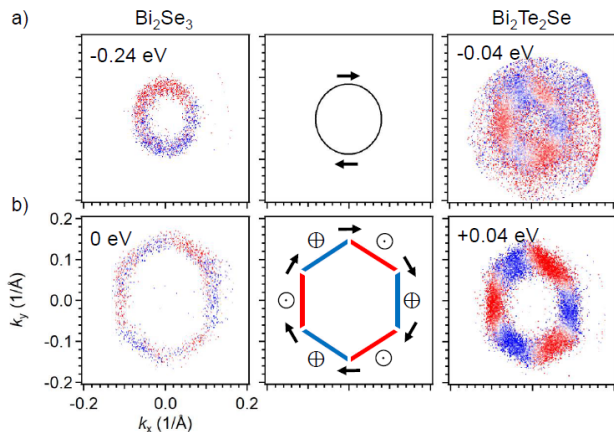


FIG. 5. Normalized CD isoenergy contours of  $\text{Bi}_2\text{Se}_3$  and  $\text{Bi}_2\text{Te}_2\text{Se}$  for energies close to (a) DP and close to (b) BCB.

defects. Moreover, the spin texture of such helical and hexagonally warped states presents an out-of-plane flipping. To investigate the CD in  $\text{Bi}_2\text{Te}_2\text{Se}$ , we measure the photoelectron intensity with  $L$ - and  $R$ -circularly polarized UV light,  $\Delta I(E, k_x, k_y, t) = (I_L - I_R)/(I_L + I_R)$ , while keeping the IR pump beam linearly polarized. Fig. 4(a) shows the normalized difference  $E - k$  cuts along  $k_x$  ( $\bar{K}-\bar{\Gamma}-\bar{K}'$ ) and  $k_y$  ( $\bar{M}-\bar{\Gamma}-\bar{M}'$ ) directions in  $\text{Bi}_2\text{Te}_2\text{Se}$  obtained by integrating  $\Delta I$  in  $k$ -space  $\pm 0.02\text{\AA}^{-1}$  around  $\bar{\Gamma}$ . Along  $k_x$ ,  $\Delta I$  flips sign whereas along  $k_y$  it is less obvious for energies inside the band-gap. This may be attributed to an in-plane and out-of-plane spin texture along  $\bar{M}-\bar{\Gamma}-\bar{M}'$  and  $\bar{K}-\bar{\Gamma}-\bar{K}'$ , respectively[9]. However, a recent spin-resolved ARPES study showed the bulk sensitivity associated to low photon energies affects the out-of-plane component of the electron spin during the photoemission process [19]. This result implies that CD studies alone do not permit to recover quantitative information about the intrinsic spin structure of the system under investigations. Looking at the entire momentum contour for DSS at various energies inside the band-gap (see Fig. 4(b)), the CD flips signs along consecutive  $\bar{K}-\bar{\Gamma}-\bar{K}'$  directions. This suggests that the out-of-plane electron spin vector along  $\bar{K}-\bar{\Gamma}-\bar{K}'$  flips sign as it varies from parallel to  $k_x$ - $k_y$  to out of the plane [19]. To certify this behavior we have also investigated the CD in  $\text{Bi}_2\text{Se}_3$  under similar geometry and experimental conditions. Similarly, Fig. 4(c) and 4(d) depict normalized  $E - k$  cuts along the high symmetry directions and momentum CD contours, respectively. The spin texture of DSS changes sign when is measured along  $k_y$  while the flip contrast along  $k_x$  is less obvious. In stark contrast to  $\text{Bi}_2\text{Te}_2\text{Se}$ , the CD sign is preserved at positive and negative  $k_y$  values for energies of DSS inside the gap. However, it gradually evolves to follow a similar trend to that of  $\text{Bi}_2\text{Te}_2\text{Se}$ . This implies that the spin texture of DSS inside the gap is purely in-plane and it is gradually deformed as energies get closer to BCB. This effect can be seen in Fig. 5(a) and 5(b) for selected

isoenergy contours inside the gap and close to BCB, respectively. On the other hand, it has been suggested that these TI possess a clear in-plane DSS spin texture for energies close to DP [37], and it should be noted that the CD-ARPES signal may present modulations depending on the excitation energy [23, 38]: in order to get a more complete description of the system, it would be thus very interesting to extend our study to various higher values of the probe photon energy.

The spin texture in DSS is further investigated as a function of the sample's azimuthal rotational angle  $\phi$ . Figure 5 shows non-normalized CD contours at  $E - E_F = +0.04$  eV in  $\text{Bi}_2\text{Se}_3$ . One can directly notice that while  $\phi$  rotates  $30^\circ$  from the initial incidence angle, the CD of DSS follows the same trend whereas that of BCB remains unchanged. Consistently with the work presented by Wang *et al.* [9] for filled electronic states, this is a clear indication that what is measured here is the real spin texture of the electronic states; the BCB states do not exhibit CD regardless of the sample rotation, while DSS do.

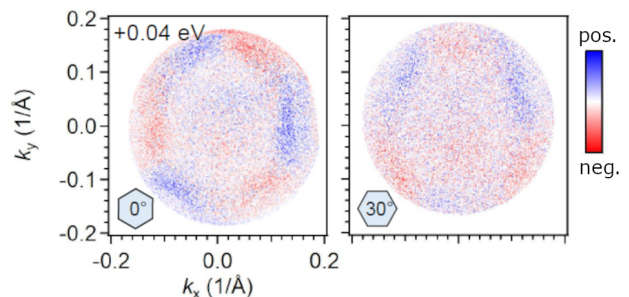


FIG. 6. Non-normalized isoenergy contours at  $+0.04$  eV for a sample rotation angle of (a)  $\phi = 0^\circ$  and (b)  $30^\circ$ , respectively. Hexagons in the inset represent the Brillouin zone of  $\text{Bi}_2\text{Se}_3$ .

#### IV. CONCLUSION

By measuring the CD of the pump-probe multi-dimensional ARPES yield, we show that the excited topological surface states of  $\text{Bi}_2\text{Te}_2\text{Se}$  present a stronger out-of-plane spin flipping with respect to  $\text{Bi}_2\text{Se}_3$ . Our instrumental configuration allows the simultaneous collection of the whole  $I(E, k_x, k_y, t)$  signal without moving the sample, making it possible to unambiguously discriminate genuine matrix element from experimental geometry effects, and represents an effective approach towards the study of spin texture in photoexcited topological materials.

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