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Time resolved ultrafast ARPES for the study of topological insulators: The case of Bi_2Te_3

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Abstract. We discuss the application of time-resolved ultrafast angle resolved photoelectron spectroscopy to the study of photoexcited topological insulators. Measurements performed on the prototype material Bi_2Te_3 clearly show that all the main processes involved in the ultrafast surface carrier dynamics of topological insulators can be clearly observed and quantitatively analyzed. The comparison with other experimental results shows that the relative position of surface and bulk conduction bands with respect to the system Fermi level play an essential role in the recombination processes following ultrafast optical excitation.

1 Introduction

Topological Insulators (TI's) are one of the topics of condensed matter physics that are currently attracting more attention [1,2]. In particular, three dimensional TI's are particularly interesting, due to their unique transport properties: they present conducting surface states, while the bulk is a semiconductor [3]. These surface states are topologically distinct from the bulk and are consequently particulary robust, due to the combined action of a strong spin orbit coupling and of time resolved symmetry. From the very beginning, Angle Resolved PhotoElectron Spectroscopy (ARPES) has played a very important role in the study of 3D TI's [4,5], since it has proven to be particulary effective in measuring the properties of the Dirac cone constituting the surface bands. Some of the properties of these Dirac cones can be studied by driving them out of equilibrium with an ultrafast optical excitation: for this kind of approach, ultrafast time resolved ARPES is a particularly effective method.

Ultrafast time resolved ARPES has emerged over the last years as one of the main techniques for the study of out of equilibrium condensed matter. As depicted in Fig. 1, it involves the use of two fs laser pulses: the first one (pump) excites

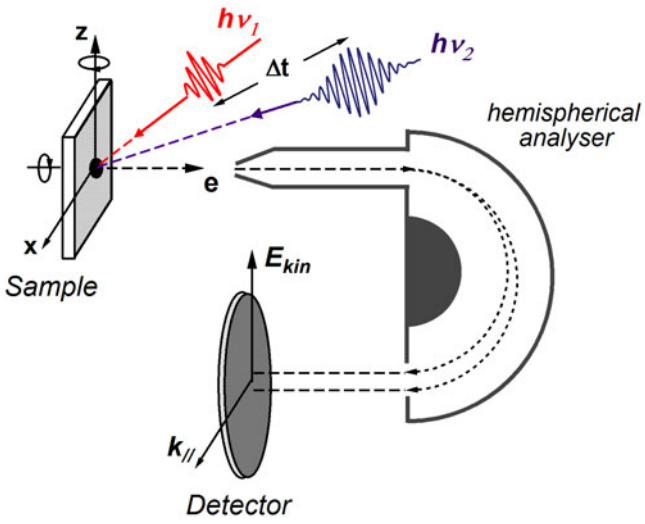

Fig. 1. Schematic representation of a pump-probe ARPES experimental setup.

electrons to a normally empty state, and the second one (probe) measures the ARPES distribution of the excited electronic structure. This technique has already provided important contributions to our understanding of various systems like for instance charge density wave compounds [6–8], Mott insulators [9,10], graphite [11] or high temperature superconductors [12,13].

In this paper we provide a clear example of application of pump-probe ultrafast ARPES on topological insulators. The distinctive features of this technique are the ones typical of ARPES (capability of measuring individual electronic bands in reciprocal space, high surface sensitivity) [14] coupled with the further advantages of a pump-probe approach (extension to the study of excited states, and to scattering and relaxation processes): for all these reasons, time resolved ARPES is particularly well adapted for the the study the dynamics of Dirac cones in TI's. The time resolved ARPES experiments so far performed on these materials have focused on the prototype 3D TI's Bi_2Se_3 [15–17] and Bi_2Te_3 [18]. In our previous work on (n-type) Bi_2Te_3 [18] we showed how the ultrafast dynamics of the surface states is affected by the scattering with the bulk conduction band; in this paper, we present an analogous study on a Bi_2Te_3 compound which is still n-type, but with a slightly different position of the Fermi level with respect to the bands [19] (within the gap rather than crossing the conduction band). Following the different scattering processes, we will show that even such small differences in the band position can affect the ultrafast evolution of the transient surface populations.

2 Experimental

Single crystals of Bi_2Te_3 were carefully oriented and cleaved under ultrahigh vacuum conditions (2×10^{-10} mbar) at 130 K. Our pump-probe photoemission measurements can be performed with an overall time resolution of 80 fs and an energy resolution of 60 meV (a detailed description of our experimental system and of its capabilities can be found in [20] and [21]). In Fig. 1, we show a schematic diagram of our experimental setup. Photoelectrons were excited from the sample with $h\nu_2 = 6.32$ eV photons, obtained as the fourth harmonic of the $h\nu_1 = 1.58$ eV laser that is used to pump the transient states in pump-probe configuration. The analyzer collects the photoelectrons

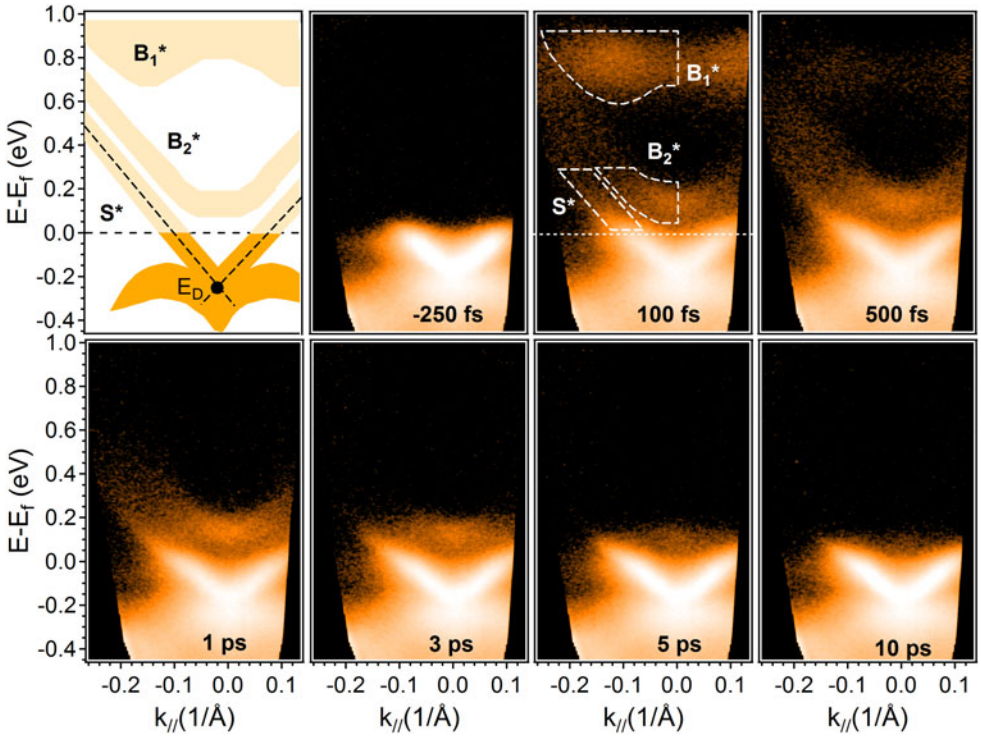

Fig. 2. Time-resolved ARPES sequence obtained after photoexcitation from the pump pulses: the color intensity is presented in logarithmic scale. In the upper left corner, a schematic view of the bands involved in the process: the bulk bands B_1^*, B_2^* and the surface Dirac Cone. In the image $\Delta t = 100$ fs the integration regions B_1^*, B_2^* and S^* are defined.

and measures both their kinetic energy E_{kin} and their momentum $k_{//}$, thus giving the evolution of different photoexcited ARPES images as a function of time delay. The use of a 6.32 eV probe gives access to a limited portion of k-space, but sufficient in this case to observe the Dirac cone which in the case of 3D TI's sits at the Γ point. Our probing depth at this low photon energy can be estimated to about (2-3) nm [22,23]: this explains why with our measurements we can optimize the detection of the surface states while still being able to observe the bulk valence and conduction bands.

3 Results and discussion

In Fig. 2, we present the evolution of the ARPES measurements obtained along the ΓM direction at different pump-probe time delays following the pump excitation (35 fs, 1.56 eV, 0.15 mJ/cm^2). In the upper left corner we present a schematic view of the bulk and surface bands as derived from theoretical calculations [3,24], to make the comparison easier with our experimental results. At negative time delays (i.e. for pump pulses arriving after the probe) the bands are in their ground state and no excitation effects can be observed. At positive time delays, different phenomena can be observed from the evolution of the various ARPES images: first the optical excitation of electrons to high energy conduction band states, then interband scattering processes

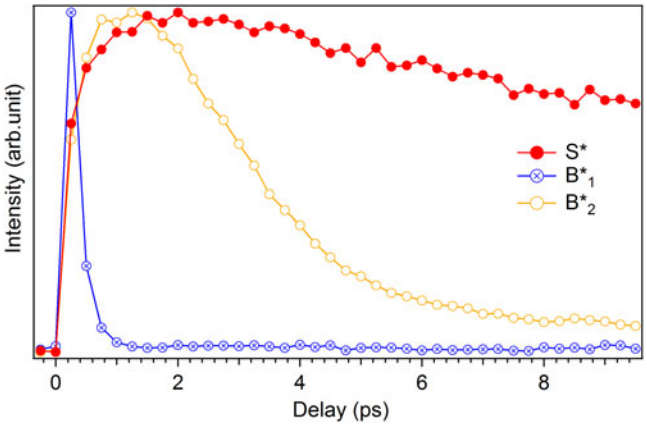

Fig. 3. Ultrafast evolution of the populations S^*, B_1^* and B_2^*.

towards low energy bulk and surface bands, followed by recombination processes [25–28] that progressively reduce the excess carrier population.

To better understand these processes, we follow as function of the time delay the evolution of the transient electron populations in different regions of reciprocal space. In Fig. 2, in the 100 fs delay window, we indicate with dashed lines three integration regions, corresponding respectively to the excited electrons in a high energy bulk conduction band (B_1^*), the low-lying bulk conduction band (B_2^*) and the Dirac cone (S^*). In Fig. 3 we show the evolution of the excited electrons in these different integration regions, as a function of time delay. One can see that the maximum of the transient electron population in the B_1^* window is at about 100 fs, earlier with respect to the other regions; subsequently, interband scattering towards the low-lying conduction band B_2^* and the Dirac cone S^* voids B_1^* at about 0.5 ps. The bulk conduction band B_2^* has a maximum population at about 1 ps and presents a strong inter-band scattering with the Dirac cone, due to the proximity of the two bands (as already pointed out in our previous work [18], the scattering rate between the two bands is faster than the time resolution of our experiment). B_2^* is almost completely relaxed at about 10 ps, giving a time scale for the main bulk recombination processes. The excess electrons in the Dirac cone (S^*) relax also through the conduction band thanks to the inter-band scattering processes: nevertheless, it is clear that the intrinsic lifetime of Dirac fermions in surface states is longer, because the excess population decreases in a markedly slower way in the Dirac cone. As already found also for other 3D TI's, this can be due to the weak electron-phonon coupling of these states [29,30]. It is worth pointing out that the time evolution curves presented in Fig. 3 are qualitatively similar to our previous work on Bi_2Te_3 [18], but all the numerical values (maxima, lifetimes) are clearly different. The main difference between the two measured surfaces is the relative position of the surface Dirac cone and bulk conduction band with respect to the Fermi level, indicating that all the described processes involved in the ultrafast dynamics of topological insulators are very sensitive to these parameters.

4 Conclusion

In conclusion, we discussed how time resolved ARPES can be a very valuable technique for the study of the transient electronic dynamics at the surface of topological insulators. Thanks to its capability of resolving individual electronic bands and to its

surface sensitivity, it is particularly well suited to observe the evolution of the surface Dirac cone. Novel instrumental solutions for the photoelectron detection, using a time-of-flight approach or efficient spin analysis [31], will probably further enhance these capabilities. Since a time resolution of 50–100 fs is nowadays easily attainable with time resolved ARPES, this method gives access to the time scale of all the main processes determining the ultrafast surface dynamics of these compounds. Based on a detailed study of the prototype material Bi_2Te_3, we discussed all these processes (ultrafast excitation, interband scattering, bulk recombination and Dirac cone relaxation), which are found to markedly depend on the position of the Fermi level with respect to the surface and bulk bands. This indicates that for a complete understanding of the non equilibrium dynamics of 3D TI's an extensive study of different materials and different surface band lineups is needed.

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