Ultrafast Electronic Dynamics of the Bi_2Se_3 Topological Insulator, revealed by Tr-ARPES

F. Cilento[1,*], A. Crepaldi[2,**], B. Ressel[1], M. Zacchigna[3], C. Grazioli[1], H. Berger[2], P. Bugnon[2], K. Kern[2,4], M. Grioni[2] and F. Parmigiani[1,5]

[1]*Sincrotrone Trieste S.C.p.A., Basovizza I-34012, Italy.*
[2]*Institute of Condensed Matter Physics (ICMP), Ecole Politechnique Fédérale de Lausanne (EPFL), CH-1005 Lausanne, Switzerland.*
[3]*C.N.R. - I.O.M., Strada Statale 14 km 163.5 Trieste, Italy*
[4]*Max-Planck-Institut (MPI) fur Festkorperforschung, D-70569 Stuttgart, Germany.*
[5]*Department of Physics, Università degli Studi di Trieste, Trieste I-34127, Italy.*

*e-mails: * federico.cilento@elettra.trieste.it; ** alberto.crepaldi@epfl.ch; webpage: www.elettra.trieste.it/labs/t-rex.html*

INTRODUCTION

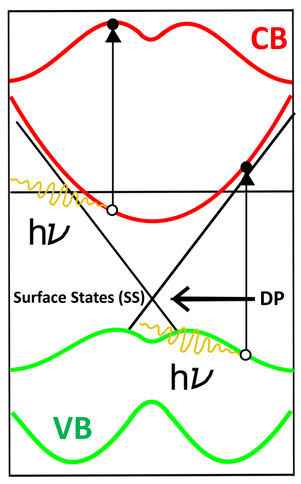
We use time- and angle- resolved photoemission spectroscopy (tr-ARPES) to reveal how the non-equilibrium electronic population in the surface state (SS) of the topological insulator (TI) Bi_2Se_3 evolves, after photoexcitation with ultrashort laser pulses.

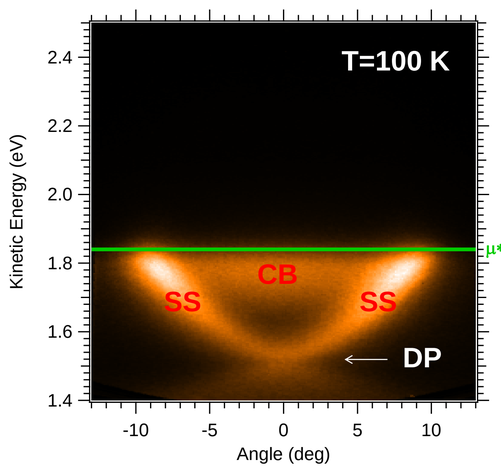
We succeed in disentangling a large increase of the effective temperature (T*) from a shift of the effective chemical potential (μ*), which is consequence of the ultrafast photodoping of the conduction band. We observe that the relaxation dynamics of T* and μ* are k-independent.

Together, these two quantities uniquely define the temporal evolution of the impulsively photoexcited charge population along the SS: we successfully modeled the relaxation dynamics of the instantly thermalized hot electrons with an effective Fermi-Dirac distribution with time-dependent temperature T*(t) and chemical potential μ*(t). We conclude that the energy dependence of the non-equilibrium charge population is solely determined by the analytical form of the effective Fermi-Dirac distribution [1].

TOPOLOGICAL INSULATORS

Topological Insulators (TI) are band insulators (semiconductors) supporting metallic spin-polarized surface states in the bulk band gap [2,3,4]. Large spin-orbit interaction induces an inversion of the energy ordering of the bands forming the gap. The opposite parities of these states requires the existence of an odd number of Dirac particles at the Fermi level. Their odd number combined with their spin polarization guarantee topological protection against backscattering [5]. This has recently motivated great effort both to elucidate the electronic properties of TIs, and to discover new TI compounds, candidates for future technological applications in spintronics.



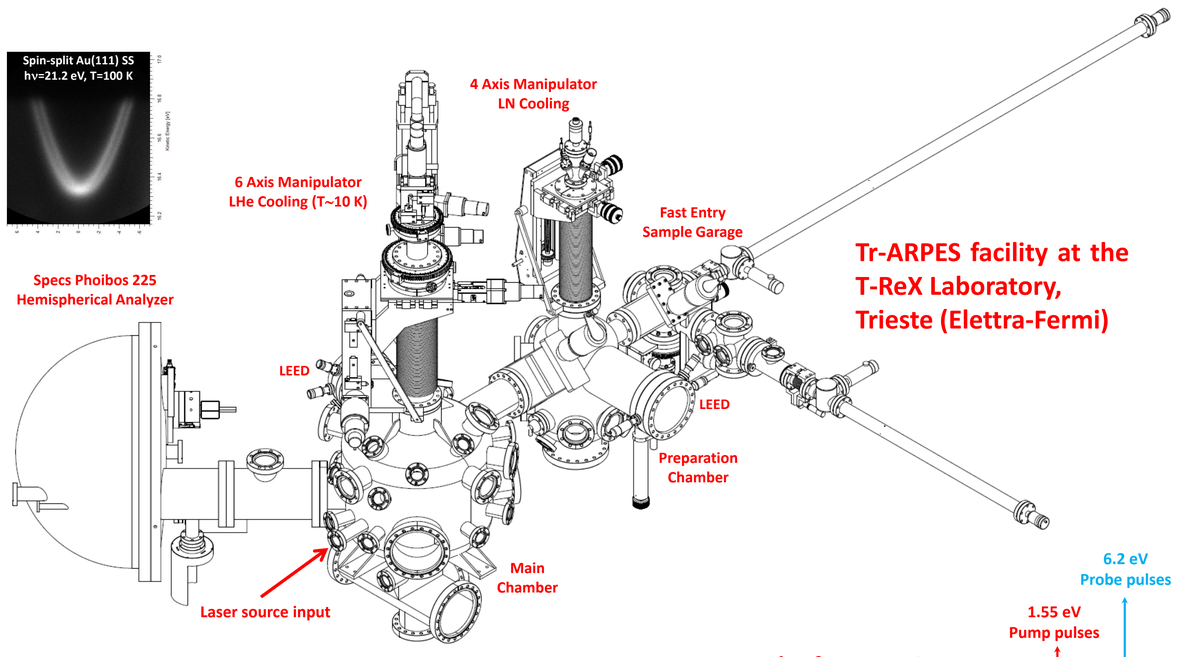


THE HIGH-RESOLUTION ARPES CHAMBER

Tr-ARPES combines the high energy and momentum resolutions of ARPES with the advantages given by a non-equilibrium approach, allowing to follow the temporal evolution of the electronic structure after the sample is brought in a non equilibrium condition by the absorption of an ultrafast laser pulse.

The non equilibrium approach allows to unravel the subtle interplay between different and often intertwined electronic or phononic degrees of freedom or collective excitations, by their different characteristic timescales and spectral features.

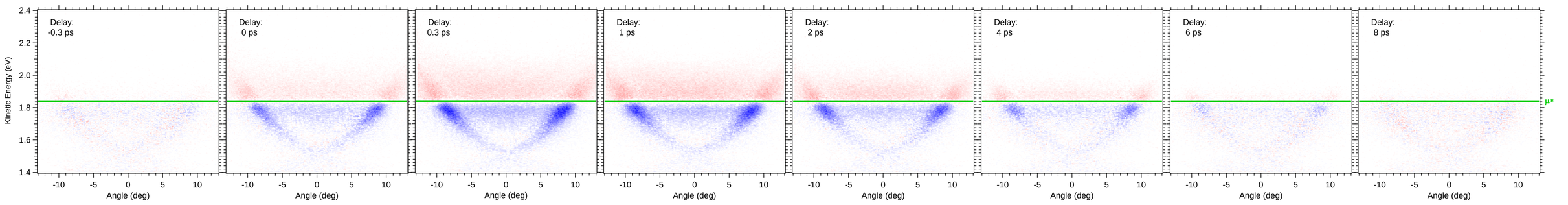
Our laboratory is equipped with a Specs Phoibos 225 electron analyzer, allowing for high energy and momentum resolutions: ~5 meV and ~0.3° are achievable. The high repetition rate ultrafast UV photon source (6.2 eV) can be tuned to choose from a high energetic or temporal resolution.



Main Features:

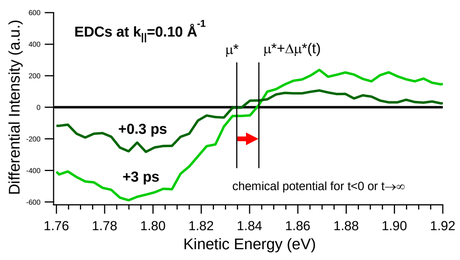
- High-Resolution Hemispherical Analyzer (ΔE~5 meV, $\Delta\vartheta$~0.3°)
- 2D TOF detector under development
- He UV Source, 21.2 eV
- LEED, Sputtering/Annealing, Cleave in <10^{-10} mbar
- Fast entry+Sample garage
- 6.2 eV Ultrafast Source (50-300 fs)
- Time-resolved ARPES with 1.55 eV photons or OPA (0.5-1.1 eV)
- Two-photon photoemission

Visualizing in real time the evolution of the transient electronic population by Tr-ARPES on Bi_2Se_3



Pump-Probe delay, t

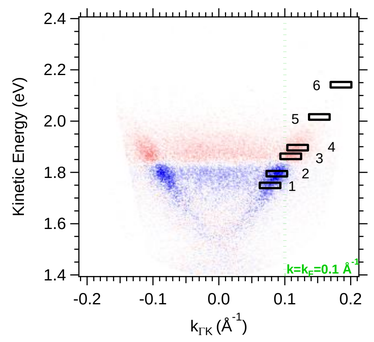
*Ultrafast shift of the effetive chemical potential, μ**



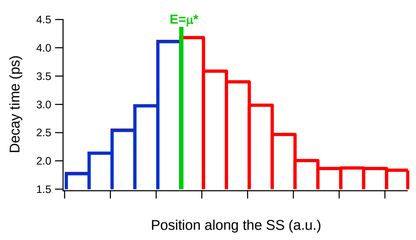
We revealed a sudden shift in the chemical potential μ* (see also [6]).

The increase in the μ* value is not compatible with the behavior predicted assuming a thermal heating for electrons in a 2D linearly dispersing band.

➔ **$\Delta\mu$* has a non-thermal origin, due to photodoping of the SS.**



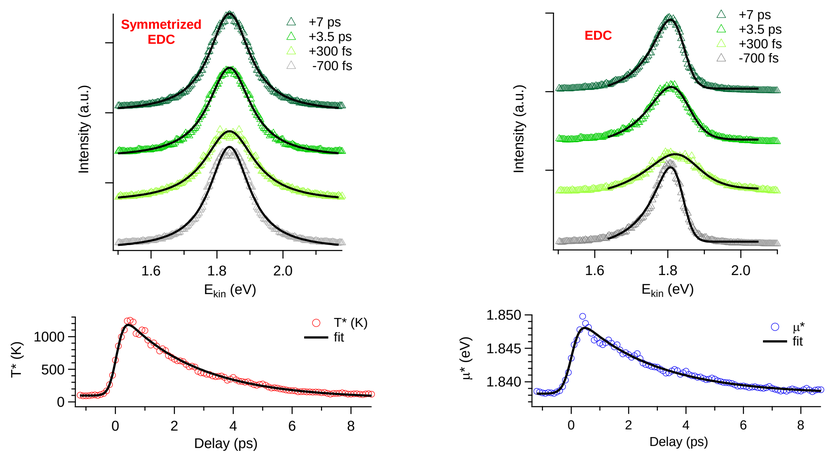
Decay-time of electrons in different small regions of (E_{kin},$k_{\Gamma K}$) space



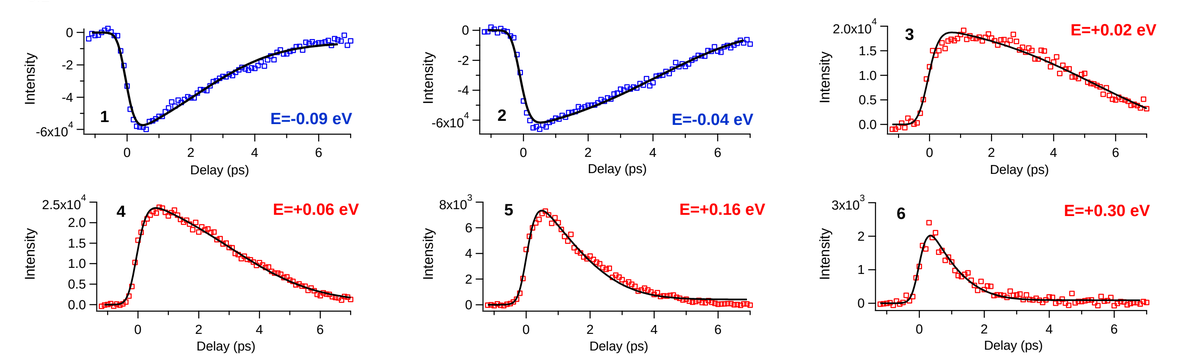
$$\zeta(E,t)=\int_{-\infty}^{+\infty}\Theta(t'-t_0)\frac{C}{1+e^{\frac{E-\mu^*(t')}{k_BT^*(t')}}}G(t-t')dt'+\phi$$

We can explain the decay time of the excited electrons, at all energies E, by assuming they obey a FD distribution with T*=T(t) and μ*=μ*(t).

Extracting T and μ* from EDCs at k=k_F*



T* and μ* relax respectively with a time constant equal to 2.5 ps and 2.7 ps. The former is compatible with a mechanism of energy relaxation to the lattice mediated by the electron-phonon interaction. The latter is related to the relaxation of the excess charge in the conduction band via diffusion.



SUMMARY

We investigated the temporal evolution of the non equilibrium electronic population in the TI Bi_2Se_3. Since at all delay times the electrons in the surface states are thermalized at an effective temperature T*, we describe their thermal distribution with an effective Fermi-Dirac distribution, where the temperature T* and the chemical potential μ* relax following an exponential decay. We clearly reveal an ultrafast modification of the chemical potential μ* ($\Delta\mu^*_{max}$~10 meV), and interpret it as the result of the ultrafast photodoping of the conduction band.

We propose a simple model, relying only on the inherent properties of the Fermi-Dirac distribution function, that fully reproduces the decay times of electrons at different points in the (E_k, $k_{//}$) space, starting from the knowledge of T*=T*(t) and μ*=μ*(t). Therfore, it provides a viable interpretation for similar energy dependence of the electron dynamics in different materials, providing a reference for future tr-ARPES studies.

1. A. Crepaldi, B. Ressel, F. Cilento et al., *submitted to PRL*.
2. L. Fu, C.L. Kane, E.J. Mele, *Phys. Rev. Lett.* **98**, 106803 (2007).
3. L. Fu, C.L. Kane, *Phys. Rev. B* **76**, 045302 (2007).
4. H. Zhang, C.X. Liu, X.L. Qi, X. Dai, Z. Fang, S.C. Zhang, *Nature Physics* **5**, 438 (2009)
5. P. Roushan, J. Seo, C. V. Parker, Y. S. Hor, D. Hsieh, D. Qian, A. Richardella, M. Z. Hasan, R. J. Cava, and A. Yazdani, Nature **460**, 1106 (2009).
6. Y. H. Wang, D. Hsieh, E. J. Sie, H. Steinberg, D. R. Gardner, Y. S. Lee, P. Jarillo-Herrero, N. Gedik, arXiv:1208.4782v1 (2012)