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The structural, chemical and surface electronic properties of $\text{Bi}_2\text{Se}_{3-y}\text{S}_y$ ($y \leq 0.66$) and $\text{Bi}_{1.993}\text{Cr}_{0.007}\text{Se}_3$

Bi_2Se_3 belongs to a class of compounds known as tetradymites, is a well-known thermoelectric material that has also come into focus in recent years for its topological insulating properties. A three dimensional (3D) topological insulator (TI) is a material that is an insulator in the bulk and has metallic conductivity at the surface. The surface conductivity of TI arises from spin-orbit interactions. Electronic states called topological surface states (TSSs) occur on the surface of TI, and exhibit linear dependence of energy on momentum [1]. TSSs are spin polarized and their backscattering from non-magnetic defects/impurities is prevented. The addition of magnetic impurities on the surface of TIs, should have an effect on TSSs, i.e., an opening of an energy gap. These properties make TIs attractive for a variety of nano electronic applications such as spin-transfer torque non-volatile memories and field-effect transistors [2], but also in the field of optoelectronics for helicity-dependent photocurrents and catalysis. Presently, material scientists are working on the synthesis and characterization of new TIs and revisiting previously known compounds from the topological insulating perspective. This is the general context of the studies reported here, where Bi_2Se_3 is the starting material. For the quasi binary system $\text{Bi}_2\text{Se}_3\text{-Bi}_2\text{S}_3$, some publications have suggested that $\text{Bi}_2\text{Se}_2\text{S}$ in its stoichiometric form should be a stable TI [3], but the few available experimental results are not yet conclusive on this issue [4,5]. We investigated the topological insulating nature of $\text{Bi}_2\text{Se}_3\text{-Bi}_2\text{S}_3$ quasi binary system for a S content in the range $0 \leq y \leq 0.66$. We prepared single crystals of $\text{Bi}_2\text{Se}_{3-y}\text{S}_y$ by the Bridgman method, study their electronic properties by means of angle-resolved photoemission spectroscopy (ARPES) and the spin-polarized ARPES. We demonstrated that the $\text{Bi}_2\text{Se}_{3-y}\text{S}_y$ solid solution supports spin-polarized TSSs for S content in the range $0 \leq y \leq 0.66$ [6].

The interest in the quasi-binary $\text{Bi}_2\text{Se}_3\text{-Cr}_2\text{Se}_3$ system stems from the search for conditions for the synthesis of Cr-doped Bi_2Se_3 bulk single crystals as an alternative to Cr-doped Bi_2Se_3 thin films [8–10]. The preparation of Cr doped Bi_2Se_3 single crystals was reported to be challenging due to the presence of secondary phase precipitates [7], while the growth of Cr doped Bi_2Se_3 thin films by molecular beam epitaxy has been reported, and the opening of the gap has been observed [8–10]. Starting from a supersaturated melt, through a process of crystallization and phase separation, we showed that is possible to prepare Bi_2Se_3 single-crystal with Cr dopant at a concentration exactly corresponding to the solubility limit without introducing secondary phases, allowing accurate measurement of ARPES maps and the observation of an apparent energy gap similarly to the case of Bi_2Se_3 thin films [11].

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