Surface doping of Bi_2Se_3(0001) by Cr and Au studied by EXAFS

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Using extended x-ray absorption fine structure (EXAFS) experiments in combination with angular resolved photoemission (ARPES) and *ab-initio* calculations we have investigated the geometric and electronic structure of ultra-thin magnetic (Cr) and non-magnetic (Au) dopant films grown by molecular beam epitaxy on the (0001) surface of the Topological Insulator (TI) Bi_2Se_3(0001). TI's host a topologically protected surface state, in which the spin is locked perpendicular to momentum making it robust against perturbations as long as time reversal symmetry is preserved. Breaking time reversal symmetry opens a gap at the Dirac point (D_P) which can be exploited to observe exotic quantum effects such as the quantum anomalous Hall effect. So far, studies focused on *bulk-doped* TI's by magnetic impurities like Cr and Mn. One complication arises from the limited saturation concentration. Several studies estimated the optimum doping level to lie in the 5 to 10% level above which a rapid deterioration of the crystallinity and an apparent reduction of the dopant's magnetic moment resulting from the formation of anti-ferromagnetically coupled metal selenides was observed. We have followed another approach by employing surface doping of Bi_2Se_3. Cr and Au were selected as a magnetic and non-magnetic dopant, respectively. Experiments were carried out at the Advanced Photon Source at the Argonne National Laboratory (USA).

In the case of Cr we find a complex evolution of the interface structure in which different adsorption sites are occupied: (i) the bismuth substitutional and the octahedral van-der Waals gap site at 0.2 monolayer coverage and in addition (ii) surface double layer formation at large film thickness (≈2.4 ML). Magnetic moments in all sites are close to $4\mu_B$ related to the Cr^{2+} species as derived by x-ray magnetic circular dichroism experiments and ab-initio calculations. Intra- and interlayer coupling is ferromagnetic and anti-ferromagnetic, respectively.

For sub-monolayer amounts of gold, we find by Au-L_{III} EXAFS experiments that even adsorption at 160 K sample temperature leads to the substitution of bismuth atoms within the topmost quintuple layer of Bi_2Se_3. Gold atoms reside in a quasi-octahedral environment of selenium atoms with distances of 2.44(3) and 2.65(7) Å indicating substantial structural relaxations upon gold incorporation as compared to the Bi-Se distances in Bi_2Se_3 (2.87 and 3.07 Å). The formation of strong Au-Se bonds results in the appearance of new gold derived d-resonance states near the Fermi level leading to an opening of a gap at the D_P of the topological surface state. Our result is a direct proof for theoretical predictions by Black-Shaffer & Balatsky [PRB85, 121103 (2012), PRB 86, 115433 (2014)].

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