Time-Resolved Spin Lifetime Measurement of Surface States on the Topological Insulator Bi2Se3

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The objective was to measure the spin lifetime of surface states on the topological insulator Bi2Se3 by a direct all-optical method. A pump-probe fs time-resolved optical study was performed on the surface of a (111) oriented Bi2Se3 sample. The sample was cleaved in air by the tape method. The 828 nm pump was incident with controlled polarization states and varied with a photoelastic modulator. The second-harmonic frequency of the probe pulse at 414 nm generated at the surface was detected with a photomultiplier and lockin detection. The sample's in-plane orientation was set to obtain surface-specific signals due to spin-polarized states. An ultrarapid transient probe SH response was observed due to the pump. The signal sign changes when the helicity of the pump and linear-polarization of the probe are reversed and provides a double-confirmation that the transient is due to spin-polarized surface carriers. The spin-lifetime is extracted from the data by curve-fitting with the measured pump-probe cross-correlation and mode-fitting to be ~50 fs. A longer transient, ~1 ps, does not depend on the helicity of the pump and is due to field-screening. Changes in field-screening after 1 hr of cleaving indicates that the surface Fermi Level increases after cleaving. 3D Topological Insulators are a new class of materials in which surface electronic states are topologically protected from backscatter: requires a spin flip. Here we have succeeded in unambiguously measuring the spin-lifetime of laser-excited carriers, albeit short. This is likely due to the surface Fermi Level being in the bulk conduction band so that pump-induced spin-polarized surface holes are rapidly filled by bulk conduction electrons. Longer spin-lifetimes will require the control of the surface Fermi Level.

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