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Evolving optical second-harmonic anisotropy at the cleaved Bi₂Se₃ surface.¹ YONG AN, AVERY GREEN, ALAIN DIEBOLD, SUNY Polytechnic Institute — Bismuth selenide (Bi₂Se₃) is a centrosymmetric topological insulator with conducting surface states. The surface states have been studied by various electrical and optical techniques in air, but ambience effects and surface aging have not been adequately addressed. Optical second-harmonic generation (SHG) is a suitable probe for the Bi_2Se_3 surface because SHG arises from symmetry breaking at the surface and thus should detect surface states preferentially over bulk states. However, a strong time dependence of SHG is often observed, hampering the detection and investigation of the surface states. Here we find a new phenomenon in which the major and minor intensity lobes of a measured rotational-anisotropy SHG pattern from a cleaved Bi_2Se_3 (111) surface can significantly change with time and eventually switch their amplitudes. This switching provides a means for tracking the progress of surface oxidation inside a quintuple layer of Bi_2Se_3 . We also perform pump-probe SHG experiments, comparatively on freshly cleaved and oxidized Bi₂Se₃ surfaces, to study charge dynamics at the oxide/Bi₂Se₃ interface and to detect spin polarization of photoexcited surface states in the Bi₂Se₃ topological insulator.

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