Optical Emission from Attosecond Tunneling Current in Bulk Solids

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Liberation of an electron from a binding atomic potential within a half-cycle of a strong optical field-tunneling ionization (TI) is a fascinating quantum-mechanical phenomenon. The attosecond temporal localization of the electron birth to the vicinity of the field peaks and the resultant sensitivity to the carrier-envelope phase of the few-cycle pulse put the TI process at the heart of attosecond science and technology. By contrast to multi-photon ionization (MPI), in which the electron plasma density increases gradually during the interaction with the optical field, the electron/ion yield in TI increases in twice-per-cycle steps, as has been recently visualized experimentally in a seminal XUV-pump-optical-probe time-of-flight spectrometry measurement [1]. The Keldysh theory of TI [2] is well developed for the tunneling from a bound atomic state into the continuum [3]. However, for inter-band transitions in condensed phase, the approach successfully used to describe TI in atomic gases encounters fundamental difficulties, such as the lack of a model for an effective electron mass in a lattice. Grasping the subtleties of the MPI and TI regimes in the ionization of solids would be of a great fundamental as well as practical importance, particularly taking into account the current prevailing view that optical damage in bulk is initiated by a high-order MPI on a defect/mimpurity.

In this work we give the first demonstration of an optical technique providing a TI signature in bulk targets, for which all types of time-of-flight electron/ion spectroscopy are inapplicable. We prove that the detected optical emission uniquely corresponds to TI and is capable of securely discriminating TI against the MPI contribution. As proposed by Brunei [4], the nearly stepwise plasma density variations at every half-cycle of the driving laser field at the frequency ω0 cause a plasma current oscillation at the frequency 2ω0. As a result, an optical field incident on this plasma is dressed with frequency sidebands spaced at multiple 2ω0 intervals. Earlier, we have reported direct observation of such multiple sidebands-tunnel-current harmonics in rare-gas targets using a non-collinear pump-probe technique [5]. Here, we report on a modified two-color technique, based on a 5-fs ionizing pump pulse at a center wavelength of 750 nm and a very weak narrowband probe pulse at 860 nm, which we have applied to record the intensity and spectral shape dependence of a TI signature around 260 nm in a number of transparent optical solids such as Fused Silica, BK7 and other glasses and isotropic and anisotropic crystals (CaF2, MgF2, Calcite). Figure 1a shows a clear 2ω0 pump-ω0 probe signature in the case of linearly polarized and mutually orthogonal pump and probe fields, whereas Figure 1b proves that this feature is completely suppressed by switching from a stepwise ionization yield to a smooth one in a circularly polarized pump field. We show that although the attosecond ionization dynamics is imprinted in the recorded spectro-temporal maps at a loss of temporal resolution and cannot be straightforwardly retrieved, nevertheless the scaling of the magnitude and shape of tunneling current emission is consistent with the known values of the tested materials’ bandgaps.

References: