High Harmonic Generation from Thin Film $LiNbO_3$

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Abstract:
We study high harmonics generation from $LiNbO_3$ films driven by femtosecond mid-infrared laser pulses. By applying an electric field, we examine changes in ferroelectric crystal symmetry influencing harmonics spectra. © 2020 The Author(s)
OCIS codes: (320.0320) Ultrafast optics; (320.7150) Ultrafast spectroscopy

High harmonic spectroscopy has been recently employed to understand the structural and symmetry properties in crystals [1]. In non-centrosymmetric solids, broken inversion symmetry results in the emission of both even and odd harmonics. Ferroelectric materials are particularly interesting, since the spontaneous permanent dipole moment can be tuned via external stimulation such as strain, applied electric fields, or THz pulse [2], and the effects may be evident in the spectral and polarization properties of emitted harmonics. Prior studies using bulk ferroelectric crystal $BaTiO_3$ indicated that high harmonics are impacted by an applied DC field [3]; however, bulk birefringence and nonlinear propagation also influence the harmonic emission. Here, we form high harmonics from thin-film $LiNbO_3$.

In the experiments, mid-infrared pulses with central wavelength of 3.4 μm, pulse energy of 10 μJ, and pulse duration of 90 fs are focused onto a 600 nm, x-cut $LiNbO_3$ thin film deposited on a fused silica substrate. The orientation-dependent spectra indicate that harmonics above 4th order exhibit a double-peak structure, as shown in Fig 1(a). Though even and odd harmonics are maximized at different crystal orientation, their local minima coincide for driving laser polarization perpendicular to the c-axis. Our results closely correspond to the HHG spectrum of odd harmonics in $BaTiO_3$ [2].

We further analyze the harmonic polarization states using a polarizer placed after the crystal. We find that the harmonics are linearly polarized, with polarization direction parallel to that of the driving laser, when the driving laser polarization is along the main symmetry axes of the crystal. However, for other orientations of the crystal, harmonic components with perpendicular polarization are also observed, as shown in Figs 1(b) and (c).

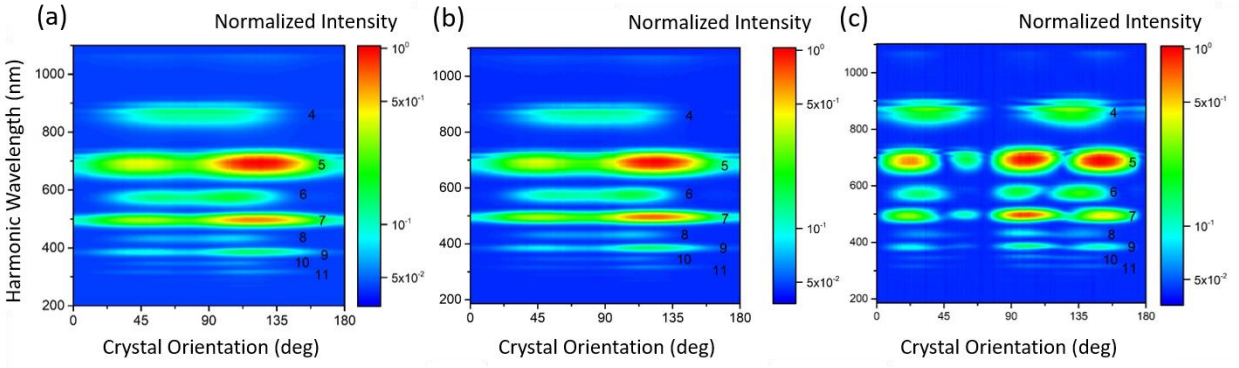

Fig. 1. Orientation-dependent harmonic spectra (a), for harmonics emitted parallel(b), and perpendicular (c) to the laser polarization

In conclusion, we demonstrate preliminary results for polarization and orientation dependent high harmonic spectrum in $LiNbO_3$ thin-films using mid-infrared laser pulses. We test potential variation in the ferroelectric crystal symmetry, which may impact both polarization and orientation dependence of generated high-order harmonics. This material is based on the work supported by the National Science Foundation under award no. 1806135 and by the Air Force Office of Scientific Research under award no. FA9550-16-1-0149.

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