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Enhancement of VUV and EUV generation by field-controlled resonance structures of diatomic molecules[1] JOHN HESLAR, National Taiwan University, DMITRY A. TELNOV, St. Petersburg State University, SHIH-I CHU, University of Kansas — Below- and near-threshold harmonic generation provides a potential approach to achieve a high conversion efficiency of vacuum-ultraviolet and extreme-ultraviolet sources for the advancement of spectroscopy. Here we perform an *all-electron* time-dependent density functional theory (TDDFT) study for the nonperturbative treatment of below- and near-threshold harmonic generation of CO and N_2 diatomic molecules subject to short near-infrared laser pulses and aligned parallel to the laser field polarization. We find that with the use of different driving laser pulse shapes we can control and enhance harmonic generation through the excited state resonance structures. Our analysis reveals several novel features where the HHG signal is enhanced, boosting the conversion efficiency on the microscopic level. Depending on the pulse shape, the enhancement can reach 5 to 7 orders of magnitude as compared to the reference sine-squared laser pulse of the same duration.

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