

Super-Harmonic Generation in Perovskite BaTiO₃: The Role of Electron-Electron Correlations

Didarul Alam

Department of Physics, University of Central Florida, Orlando, FL 32816

Didarul.alam@ucf.edu

Higher-harmonic generation (HHG) is a powerful experimental technique for studying static and dynamic properties of electron systems in solids like [1,2]. However, the role of correlation effects in the higher harmonic (HH) spectrum is still not well understood. We analyzed how electron-electron correlations affect the HH spectrum in perovskite BaTiO₃ excited by a laser pulse by applying ab initio time-dependent density-functional theory. To accurately take into account the effects of electron-electron correlations we used the exchange-correlation (XC) kernel obtained from the charge susceptibility of the effective Hubbard model solved with the Dynamical Mean-Field Theory [3,4]. As we have found, correlation effects significantly change the interband current (left Figure 1) and shift the HH spectrum to higher frequencies (right of Figure 1). In addition, it was found that when the correlation strength (magnitude of the XC kernel) exceeds a critical value, the HH spectrum displays super-harmonics - periodic enhancement of equally-distanced harmonics (the step proportional to the correlation energy) and suppression of the harmonics between them (e.g. harmonic order 4, 8, 12 so on).

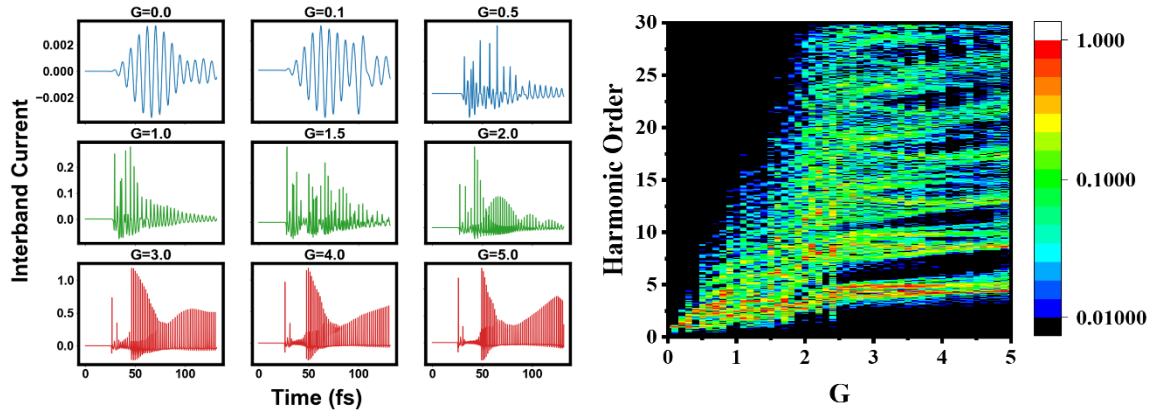


Figure 1: (left) Interband current, (right) harmonic spectrum for different values of correlation for pulse duration 68 fs, field strength $E_0 = 0.1 \text{ V/\AA}$ and pulse frequency $\omega_0 = 0.5 \text{ eV}$.

Our results, valid for a wide range of pulse parameters, can be used to quantify the role of correlation effects in perovskites and other strongly correlated materials [5].

References

- [1] S. Ghimire, A. D. DiChiara, E. Sistrunk, P. Agostini, L. F. DiMauro, and D. A. Reis, Observation of High-Order Harmonic Generation in a Bulk Crystal, *Nature Physics* **7**, 2 (2011)
- [2] S. Ghimire and D. A. Reis, High-Harmonic Generation from Solids, *Nature Physics* **15**, 1 (2019).
- [3] V. Turkowski and T. S. Rahman, Nonadiabatic Exchange-Correlation Kernel for Strongly Correlated Materials, *Journal of Physics: Condensed Matter* **29**.45 (2017).
- [4] D. Alam, N. Ud Din, M. Chini, and V. Turkowski, Electron-Electron Interactions and High-Order Harmonics in Solids, *Phys. Rev. B* **106**, 235124 (2022).
- [5] D. Alam, M. Chini, and V. Turkowski, Electron Correlations and Memory Effects in High Harmonic Generation from Perovskite BiFeO₃, *Conference on Lasers and Electro-Optics* (2023)