Ultrafast Optical Nonlinearity of Transparent Conducting Oxides

Y Sivan[1]

[1] *Electrical and Computer Engineering, Ben-Gurion University of the Negev, Be'er Sheva, Israel*
Contact Email: sivanyon@bgu.ac.il

The exploration of light-matter interactions using intense, extremely short optical pulses have been receiving growing interest in the fields of condensed matter and photonics, as they are key to realizing exotic light manipulations such as time-reversal (aka time reflection) and photonic time crystals. These effects require unusually large (order unity) permittivity changes, occurring on a sub-cycle ("instantaneous") time scale.

While most conventional nonlinear optical materials provide either a strong or instantaneous permittivity changes, transparent conducting oxides (TCOs) have recently emerged as possible candidates allowing one to enjoy the best of these two worlds [1, 2]. Initial experimental studies relied on qualitative phenomenological or macroscopic thermal models, providing valuable insights but lacking quantitative match. To bridge this gap, our early theoretical works [3, 4] provided a comprehensive quantitative description of the electronic, thermal, and optical response of TCOs. However, the approach used is formally unsuitable to describe correctly the dynamics under intense sub/single-cycle illumination, essential for realizing time-reversal and photonic time crystals.

To close this knowledge gap, we developed a suitable density matrix formulation that provides a comprehensive description of the plethora of effects emerging under these conditions. Specifically, we unveil coherence-induced doubled frequency oscillations in the electron distribution, transient inversion, stimulated emission and excited-state absorption. We also characterize the unusual thermalization and relaxation, providing a simple theory that could be useful for the investigation of far-from-equilibrium electron dynamics in other systems. Finally, we comment on the possibility to realize time-reversal and photonic time crystals in TCOs.

References

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