Toward high-harmonic spectroscopy of strongly correlated electron systems

Yuta Murakami

Institute for Materials Research, Tohoku University, Japan

CEMS, RIKEN, Japan

Email: yuta.murakami@tohoku.ac.jp

The development of recent laser technologies opens new avenues for nonlinear spectroscopy as well as various routes to the ultrafast manipulation of quantum materials. High-harmonic generation (HHG) is a fundamental nonlinear optical phenomenon arising from strong light–matter interactions. Initially discovered in gases, the concept of HHG has recently been extended to condensed matter systems. While most studies have focused on weakly correlated band insulators, increasing attention is now directed toward strongly correlated electron systems (SCESs). SCESs provide a new frontier for HHG research owing to their nontrivial excitation structures, rich phases, and their dynamical tunability [1].

In this talk, we discuss the physics of HHG in SCESs and its potential spectroscopic applications, with a particular focus on Mott insulators—prototypical SCESs governed by strong Coulomb interactions.

First, based on a series of theoretical analyses of the Hubbard model with different dimensionalities, we establish the fundamental picture of HHG in Mott insulators and clarify the crucial role of spin–charge coupling [2–4]. HHG in these systems can be understood within a three-step framework describing the nonlinear dynamics of charged elementary excitations emerging from strong correlations. The nature of these elementary excitations depends sensitively on dimensionality due to the spin–charge coupling. We illustrate this using the two-leg ladder system [4], where the coupling gives rise to characteristic dephasing processes and fractionalization of the HHG signal.

Second, we demonstrate that HHG in SCESs can serve as a probe of ultrafast evolution of electronic states during photoinduced phase transitions [5]. Because SCESs are sensitive to laser excitation, the strong fields driving HHG can simultaneously trigger phase transitions. In Sr_2CuO_3, we observe redshifts of high-harmonic peaks associated with a photoinduced insulator–metal transition. Using nonequilibrium dynamical mean-field theory, we show that these redshifts originate from ultrafast modifications of the electronic structure, which alter charge dynamics cycle-by-cycle.

Our results underpin the intriguing features of HHG in SCESs and pave the way for its potential applications in ultrafast spectroscopy.

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