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HIGH-ORDER HARMONIC GENERATION IN SOLIDS

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Foreword

The first experiments on the observation of non-perturbative high-order harmonics from solid materials subjected to strong laser fields were conducted by Shambhu Ghimire, David A. Reis and their collaborators and the results reported in a *Nature Physics* letter in 2010. Before this, experimental studies of strong field-driven high-order harmonic generation (HHG) phenomena were focused on isolated atoms and molecules in the gas phase. The challenges of intense laser-matter interactions in condensed matter lie in plasma formation and subsequent sample damage. In Ghimire *et al.*'s experiments, these concerns were mitigated by the tight focusing of laser beam to the exit face of 0.5 mm thick zinc oxide (ZnO) crystals. The ZnO crystal has a band gap of 3.2 eV and the driving laser's photon energy was 0.3 eV, making the band gap to photon energy ratio $\sim$10, which is the same as the ratio of the ionization potential of argon atom (15.6 eV), the mostly used gaseous target, to the photon energy of the most common laser (1.5 eV) used in those experiments. Obtaining such a large ratio was important to ensure that multi-photon absorption is suppressed, and the interaction is well into the strong-field tunneling regime. In these experiments, the samples did not get damaged and there was no sign of plasma formation up to the peak intensity $\sim 5 \times 10^{12}$ W/cm^2 for repetitive excitation at 1 kHz. High-order harmonics up to 25$^{\text{th}}$ orders were observed, and the spectrum included a plateau feature, and an abrupt high-energy cutoff, very much like in atomic and molecular HHG. However, the high-energy cutoff law $I_p + 3.17\,U_p$, did not hold in solid-state HHG. The cutoff scaled linearly with the peak field of the laser instead of the quadratic scaling that has been well established in gaseous media. This new scaling hinted at a novel microscopic mechanism in solids attributed to the fundamental role of high density and periodicity. This paper sparked interest in the attosecond science community with several open questions at the time: (i) what are the relative roles of inter-band and intra-band channels, (ii) is there a unique relation between high-harmonic spectra and electronic band structure of the source material, (iii) can solid-state HHG be a novel probe of the atomic scale structure and dynamics of the source material, and (iv) can solid-state HHG support coherent attosecond pulse generation? Now it is well over a decade since the first experiments and these questions are at least partially answered, as presented beautifully in this book, and with a lot of exciting experimental and theoretical works ahead of us.

Shambhu Ghimire
David A. Reis

Dr. Shambhu Ghimire is a Lead Scientist at Stanford PULSE Institute, SLAC National Accelerator Laboratory, Stanford University, USA. He is a Fellow of OPTICA (formerly Optical Society of America), and is known for his pioneering work in generation of isolated attosecond pulse using polarization gating, strong-field physics in bulk and 2D crystals, nonlinear X-ray optics, and solid-state high-harmonic generation.

Dr. David Reis is the Director of Stanford PULSE Institute and is a Professor of Photon Science and Applied Physics at SLAC National Accelerator Laboratory and Stanford University, USA. He is a Fellow of OPTICA (formerly Optical Society of America) and the American Physical Society, and is known for his pioneering work in ultrafast and strong-field science including non-equilibrium lattice dynamics measurements, nonlinear X-ray optics, and solid-state high-harmonic generation.

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Preface

High-order harmonic generation (HHG) in solids represents a captivating phenomenon at the intersection of attosecond science and condensed matter physics. Unlike the more commonly studied HHG in gases, which involves the interaction of intense laser fields with individual atoms or molecules, HHG in solids occurs when strong laser pulses interact with a crystalline lattice, leading to the emission of high-energy photons at harmonics of the driving laser frequency.

The phenomenon of HHG in solids involves both the acceleration of electrons within the crystal lattice and the electron-hole dynamics between the conduction and valence bands, both processes driven by the intense laser field. As these electrons gain energy from the laser, they undergo rapid oscillations within the lattice structure or transitions between the conduction and valence bands, emitting high-energy photons. This results in the generation of an attosecond burst of coherent radiation, enabling scientists to probe ultrafast dynamics in solid-state systems with unprecedented temporal resolution, as well as to characterize the crystal structure with sub-angstrom spatial resolution.

The Nobel Prize in Physics in 2018 was awarded to Gérard Mourou and Donna Strickland for their work on high-intensity ultrashort laser pulses. The techniques and principles they developed for the generation of intense laser pulses have paved the way for advancements in HHG research, including applications in solid-state systems.

Last year, the 2023 Nobel Prize in Physics was awarded to three trailblazers of attosecond science: Pierre Agostini, Ferenc Krausz, and Anne L'Huillier. The Laureates' experiments have produced pulses of light so short that they are measured in attoseconds, thus demonstrating that these pulses can be used to provide real-time images of processes inside atoms and molecules, and, recently, solid crystals.

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By pushing the boundaries of our understanding of light-matter interactions in solids, HHG continues to unlock new frontiers in ultrafast science, offering unprecedented insights into the dynamics of electrons in condensed matter systems and laying the groundwork for future advancements in fields ranging from materials science to quantum electronics. The intent of this book is to provide a glimpse of recent theoretical and experimental developments in this research area.

We owe a great debt of gratitude to all of the authors, who have made this endeavor possible. We are especially grateful to Shaun, from World Scientific Publishing Co. Without him, this book would not be a reality. We hope that the present book serves as a valuable reference resource for many years to come.

Sincerely,

Marcelo Ciappina
Paraskevas Tzallas
Editors

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About the Editors

DSc. Dr. Marcelo Ciappina is Associate Professor at the Guangdong Technion–Israel Institute of Technology, China. He is also Guest Professor at Nanjing University of Science and Technology, China, and Associate Editor of *Ultrafast Science*, a *Science* partner journal. He has co-authored over 250 journal papers with an H-index of 34. He is regarded as an expert in theory and numerical simulations of nonlinear laser interactions with atoms, molecules and complex systems. In recent years he has been one of the pioneers in a novel and fascinating field that merges two relatively new areas of research: attosecond and nanoscale physics.

Dr. Paraskevas Tzallas is Research Director at the Foundation for Research and Technology — Hellas (FORTH), Heraklion (Crete), Greece, Head of the Attosecond Science and Technology activity at FORTH, and Scientific Advisor of secondary sources at Extreme Light Infrastructure-Attosecond Light Pulse Source (ELI-ALPS), Szeged, Hungary. His research interest is in the field of attosecond science and quantum light engineering for applications in quantum information science. His research achievements, among others, include the first direct observation of attosecond light bursts emitted in gas and solid-state media, as well as the first observation of optical Schrodinger "cat" states using intense laser-matter interactions.

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