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Annkatrin Madlen Sommer

Ultrafast Strong Field Dynamics in Dielectrics

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Ultrafast Strong Field Dynamics in Dielectrics

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Supervisor's Foreword

Progress in science and technology during the last century provided access to fast and cost-effective computing. Until today, the continuous downscaling of semiconductor electronic devices has allowed to double the computing performance of integrated circuits every second year, as predicted by Gordon E. Moore in 1965. On the one hand, the clock rate of transistors increases inversely to their size; on the other hand, more devices can be incorporated into a single microprocessor chip. Nowadays, devices with a spatial extent of 20 nanometers can be fabricated and operated up to clock rates of 10 gigahertz in contemporary digital integrated circuits. However, further substantial improvement in processing speed appears to be prevented by heat generation, and calls for a new paradigm in electron-based signal manipulation.

Advances in solid-state laser technology over the last decade have enabled the generation of intense few-cycle optical pulses. Substantial confinement of electromagnetic energy to a single wave cycle has allowed transparent matter to be exposed to unprecedented field strengths in a reversible fashion. This has prompted the question whether strong light fields can be used for near-dissipation-free manipulation of the electronic properties of solid matter. If this question can be answered in the affirmative, it might open the door for optical signal modulation and switching of up to multi-THz frequencies.

To determine the feasibility of such an all-optical approach, the nonlinear interaction of ultrashort strong fields and matter has to be understood in more detail. Since electron dynamics in solids evolve on an attosecond timescale, a corresponding temporal resolution is required to investigate light-field-driven electron dynamics in solids directly in the time domain. The work presented in this thesis draws on major tools and concepts of attosecond metrology to perform time-resolved spectroscopy of strong-field-induced nonlinear electron dynamics. Several experimental techniques have been used to systematically investigate electron processes underlying the response of wide-gap dielectrics to the ultra-strong electric field of few-cycle near-infrared laser pulses. These developments culminate in the demonstration of a truly innovative experimental concept

dubbed attosecond polarization spectroscopy. This technique permits, for the first time, tracking of the attosecond nonlinear polarization response of matter to optical fields, thereby providing direct access to the field-matter energy transfer dynamics with sub-cycle resolution. The concept is based on the key notion of comparing the waveforms of a few-cycle laser pulse transmitted through a thin sample at low and high intensity under otherwise identical experimental conditions. Both waveforms are recorded with an attosecond streak camera. The comparison eliminates linear dispersive effects and permits determination of the oscillating nonlinear polarization response of the sample. This, in turn, yields the energy density transferred from the field to the electronic systems and vice versa, accurately quantified and time-resolved on an attosecond scale.

The energy transfer dynamics can be interpreted in terms of virtual and real population transfer into the conduction band, being responsible for the reversible modification of the electronic properties of the sample and concomitant heat dissipation, respectively. Most importantly, the work identifies a parameter range in which the former effect transiently alters the materials optical and electronic properties significantly, with the latter effect causing immeasurably small dissipation. In addition, this method, together with an elaborate theoretical concept, allows us to record the buildup of the nonlinear polarization wave in the laser ionization of neon. The increasing degree of ionization throughout the laser pulse intensity envelope yields an increasing negative contribution to the refractive index, observed as a growing phase advance of the field cycles of the strong laser pulse compared to those of the weak reference pulse. The evaluated nonlinear polarization yields a textbook example for the insight the new method provides into atomic-scale electronic motion. As a matter of fact, the measured polarization can be interpreted in terms of electrons preferentially liberated near the maxima of the electric field oscillations, performing oscillations shifted by half a wavelength compared to the driving field.

The thesis 'Ultrafast Strong Field Dynamics in Dielectrics' represents a major advance in the field of ultrafast spectroscopy. Already, the first application of attosecond polarization spectroscopy has provided a wealth of new insight into electron phenomena of fundamental and potentially great technological importance. The exploration of the ultimate frontiers of electron-based signal processing and viable routes to approaching those frontiers may benefit from these advances. A reading is highly recommended to all those working at the forefront of ultrafast science.

Garching, Germany
April 2016

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List of Publications and Conference Contributions

- **Sommer**, E. M. Bothschafter, S. A. Sato, C. Jakubeit, T. Latka, O. Razskazovskaya, H. Fattahi, M. Jobst, W. Schweinberger, V. Shirvanyan, V. S. Yakovlev, R. Kienberger, K. Yabana, N. Karpowicz, M. Schultze, F. Krausz, *"Attosecond nonlinear polarization and light-matter energy transfer in solids."*, **Nature** 534, 86–90 (2016).
- F. Habel, V. Shirvanyan, M. Trubetskov, C. Burger, **A. Sommer**, M. F. Kling, M. Schultze, V. Pervak, *"Octave spanning wedge dispersive mirrors with low dispersion oscillations"*, **Optics Express** 24(9), 9218–9223 (2016).
- M. Schultze, E.M. Bothschafter, **A. Sommer**, S. Holzner, W. Schweinberger, M. Fiess, M. Hofstetter, R. Kienberger, V. Apalkov, V. S. Yakovlev, M. I. Stockman, F. Krausz, "*Controlling dielectrics with the electric field of light.*", **Nature** 493, 758 (2013).
- W. Schweinberger, **A. Sommer**, E. M. Bothschafter, J. Li, F. Krausz, R. Kienberger, M. Schultze, "*Waveform-controlled near-single-cycle milli-joule laser pulses generate sub-10nm extreme ultraviolet continua*", **Optics Letters** 37(17), 3573–3575 (2012).
- **Sommer**, E. M. Bothschafter, S. A. Sato, C. Jakubeit, T. Latka, O. Razskazovskaya, H. Fattahi, M. Jobst, W. Schweinberger, V. Shirvanyan, V. S. Yakovlev, R. Kienberger, K. Yabana, N. Karpowicz, M. Schultze, F. Krausz, "*Attosecond Spectroscopy of Nonlinear Polarization Dynamics.*", Conference on Ultrafast Optics, Beijing (China) 2015.
- M. Schultze, E. M. Bothschafter, **A. Sommer**, S. Holzner, M. Fiess, M. Hofstetter, R. Kienberger, V. Apalkov, V. S. Yakovlev, M. I. Stockman, F. Krausz, "*Strong-field-induced attosecond dynamics in SiO_2.*", EPJ Web of Conferences 41, 02014 (2013).

Abstract

Owing to the progress in laser technology in the last decades, stable and reliable sources for ultrashort light pulses are currently available (Brabec and Krausz Rev Mod Phys 72:545–591, 2000). These pulses contain only a couple of field cycles and possess a duration of a few femtoseconds at optical frequencies. If they were used in signal processing the clock frequency could be increased by more than a factor of 1000 compared to current electronic semiconductor technology (Caulfield and Dolev Nature Photon 4:261–263, 2010). The work presented in this thesis examines whether the ultrafast strong field dynamics in dielectrics can be exploited for such an application. For this purpose it is necessary to understand the physical origin of the different dynamics triggered in the light–matter interaction on femtosecond timescales.

If an electric field interacts with matter it excites oscillating dipoles which themselves emit a polarization wave. As long as the driving field is weak the dipoles follow the external force linearly. When the amplitude of the strong field becomes comparable to the interatomic forces, the material response contains a nonlinear contribution. This nonlinear polarization includes all the characteristics of the nonlinear strong field interaction. It provides information about the response time of the system and the energy transfer dynamics throughout the interaction. The aim of this thesis is to extract the nonlinear polarization wave induced by optical few-cycle fields in wide-gap solids.

To do so, the nonlinear interaction of intense sub-4-femtosecond (*fs*) light pulses with insulators close to optical breakdown is investigated with different experimental approaches. In a first step the nonlinear Kerr coefficient is determined with time integrating techniques. The obtained result is 23 times smaller than reference values reported in the literature for the interaction with longer pulses Milam (Review and Assessment of Measured Values of the Nonlinear Refractive-Index Coefficient of Fused Silica. Appl Opt 37:546, 1998). To understand this deviation the induced material response has to be examined with a time-resolved measurement. As the electron dynamics in solids evolve on sub-femtosecond timescales, extreme ultraviolet (XUV) radiation with a few 100 attosecond (*as*) pulse duration provides the required temporal resolution. In the first experimental approach,

the modulation of the absorptivity of these attosecond pulses in the presence of the strong field is investigated. The examination reveals that the strong field triggers dynamics which can follow the driving force reversibly on sub-femtosecond timescales. In an additional nonlinear autocorrelation measurement, the highly nonlinear increase of the strong field reflectivity at intensities close to optical breakdown is observed.

To get a more comprehensive picture of the underlying dynamics which modify the XUV absorptivity and the amplitude of the reflected strong field, the experimental technique of 'Attosecond Polarization Spectroscopy' (APS) is developed. APS is an advancement of conventional pump-probe spectroscopy as it possesses attosecond resolution independent of the temporal extent of the probe pulse. It provides access to the complete nonlinear modification of the strong field throughout the light–matter interaction. As the experimental observables are analytically related to the nonlinear polarization, its amplitude and phase is directly accessible.

To verify the novel concept, APS is used to investigate the field-induced ionization process and plasma dynamics in gas. The characteristic signature of the generated plasma can be detected in the modification of the optical material density. The nonlinear response of the medium is found to be dominated by the acceleration of the freed electrons in the field. The experimental results directly reveal that the excited electron wave packet oscillates on length scales of 2–3 *nm*.

In the examination of the ultrafast strong field dynamics in dielectrics with APS, an increase of the optical material density in the presence of the strong field is observed. The modification is reversible on femtosecond timescales. Since APS provides access to the nonlinear polarization, the physical mechanisms underlying the nonlinear process can be revealed. The generation of virtual conduction band population (Yablonovitch et al. Phys Rev Lett 63(9):976–979, 1989) is identified as the source of the refractive index change. When the field strength increases at the beginning of the pulse, energy is transferred from the field to the material to move electrons away from their equilibrium positions. The projection of the laser dressed populated valence band states onto the empty conduction band states generates excited virtual carriers. As the field strength decreases the major part of the system returns to its undistorted initial state. The excited electrons decay via stimulated emission and energy flows back to the field. Only a small fraction of real carriers is excited into the conduction band throughout the interaction. Their negative contribution to the optical material density is not detectable in the experiment. The generated real conduction band population does not return to the valence band in the presence of the laser pulse. The carriers cannot return their excitation energy back to the field and therefore determine the energy irreversibly dissipated in the interaction.

The amount of energy transferred in the interaction influences the response time of the electronic system to the driving force. As absorption increases with the amplitude of the external field, the material response is decelerated with growing field strength. At intensities close to optical breakdown an upper limit of <100 *as* is extracted for the electronic response time of the system.

The maximum number of virtual carriers was generated by the strong field scales with the square of its intensity as predicted by the Kerr effect. Since the presence of virtual conduction band population increases the optical density of the medium, the amplitude of the nonlinear refractive index change at the peak of the field envelope also follows the Kerr prediction. It grows linearly with intensity without any sign of saturation up to critical field strength. The temporal evolution of the refractive index change throughout the interaction cannot be described exclusively by the Kerr effect. Its sophisticated time structure is caused by the interplay between nonlinearity and dispersion for ultrashort broadband light pulses.

The manipulable refractive index could be used as a signal in future ultrafast signal processing. The real charges generated in the interaction are excited via multiphoton absorption. For wide-gap solids and optical fields this means that their number increases with the sixth power of the field intensity. The different scaling of the generation mechanism of virtual and real carriers with intensity can be used to maximize the ratio between the induced change in optical density and the energy dissipated in the nonlinear interaction. It can be exploited to optimize the parameters of a future optical switch operating at clock rates above 100 terahertz (*THz*). By reducing the signal to the smallest detectable level, the energy density dissipated in one switching cycle can be reduced several orders of magnitude below the characteristic parameters of state-of-the-art gigahertz (*GHz*) electronic transistors (Taur and Ning, Fundamentals of Modern VLSI devices, 2nd edn., 2009).

The results obtained from the APS experiments show that the optical material density of dielectrics can be reversibly manipulated with ultrashort strong fields on femtosecond timescales. The medium reacts fast enough to follow fields oscillating at petahertz (*PHz*) frequencies. The amount of energy dissipated in the interaction is effectively tunable with the intensity of the driving field. These findings prove that the modulation of the optical properties of dielectrics with few femtosecond strong fields is a valid approach for future ultrafast signal processing.

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