
BestMasters

Springer awards „BestMasters“ to the best master’s theses which have been completed at renowned universities in Germany, Austria, and Switzerland.

The studies received highest marks and were recommended for publication by supervisors. They address current issues from various fields of research in natural sciences, psychology, technology, and economics.

The series addresses practitioners as well as scientists and, in particular, offers guidance for early stage researchers.

Johannes Schötz

Attosecond Experiments on Plasmonic Nanostructures

Principles and Experiments

With a Preface by Prof. Dr. Matthias Kling



Springer Spektrum

Johannes Schötz
Garching, Germany

BestMasters

ISBN 978-3-658-13712-0

ISBN 978-3-658-13713-7 (eBook)

DOI 10.1007/978-3-658-13713-7

Library of Congress Control Number: 2016937504

Springer Spektrum

© Springer Fachmedien Wiesbaden 2016

This work is subject to copyright. All rights are reserved by the Publisher, whether the whole or part of the material is concerned, specifically the rights of translation, reprinting, reuse of illustrations, recitation, broadcasting, reproduction on microfilms or in any other physical way, and transmission or information storage and retrieval, electronic adaptation, computer software, or by similar or dissimilar methodology now known or hereafter developed.

The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.

The publisher, the authors and the editors are safe to assume that the advice and information in this book are believed to be true and accurate at the date of publication. Neither the publisher nor the authors or the editors give a warranty, express or implied, with respect to the material contained herein or for any errors or omissions that may have been made.

Printed on acid-free paper

This Springer Spektrum imprint is published by Springer Nature
The registered company is Springer Fachmedien Wiesbaden GmbH

To my family, friends and colleagues

Preface

Attosecond nanophysics is a new research field merging ultrafast science with time scales reaching into the attosecond domain with studies on nanoscale materials. An attosecond is incredibly short. To put it in perspective, one attosecond (one attosecond = 10^{-18} seconds) compares to one second roughly as one second compares to the age of the universe. Within one attosecond even light only travels a distance of 0.3 nanometer (1 nanometer = 10^9 meter). Attosecond time and nanometer length scales are thus inherently connected. The attosecond timescale is particularly important for electrons, which are light enough to move so fast that they must be clocked with attosecond precision to track their motion. These fast electron dynamics govern the interaction of light with matter and form the basis for optoelectronics. The possibility to steer electronic processes in nanomaterials with tailored lightwaves can be exploited in ultrafast nanoelectronic circuitry with switching frequencies approaching the petahertz domain (many orders of magnitudes above conventional electronics). This potential has motivated the rapid growth of attosecond nanophysics.

The master thesis of Johannes Schötz discusses an important experimental advance in this young field, namely the ability to measure the evolution of fields on the nanoscale in real-time, i.e. attosecond timescales. He describes both experimental and theoretical advances towards the realization of the attosecond streak-camera technique on the nanoscale. While the attosecond streak-camera has become a standard tool in attosecond physics, and related measurements on electron dynamics in atoms, molecules, and extended surfaces, its realization for measurements of nanostructures is not straightforward.

The reasons are discussed in detail in the thesis, with a special emphasis on metallic nanotips, which Johannes Schötz has investigated in his work.

In attosecond streaking, electrons are photoemitted through an attosecond light pulse in the extreme ultraviolet and are accelerated by an external field provided by e.g. a synchronized optical light pulse (with a duration of a few cycles). While in conventional attosecond streaking the external fields are spatially homogenous, the near-fields of nanostructures are inhomogenous. The ramifications of the nanometer spatial inhomogeneity are non-trivial and therefore typically detailed simulations of the streaking process and its application in real-time measurements of nanoscale near-fields are required. Johannes Schötz performed such simulations and shows them in his thesis.

The thesis not only describes the first steps into the new territory of attosecond resolved measurements on nanostructures, but it is also written such that it provides guidance to a newcomer. The thesis of Johannes Schötz is of high relevance to future research in attosecond nanophysics and I wish that his ground breaking work will find the wide and interested readership that it certainly deserves.

Prof. Matthias Kling

Ultrafast Nanophotonics Group,
Laboratory of Attosecond Physics
Department of Physics, Ludwig-Maximilians-Universität München &
Max Planck Institute of Quantum Optics, Garching, Germany

The Laboratory of Attosecond Physics (LAP)

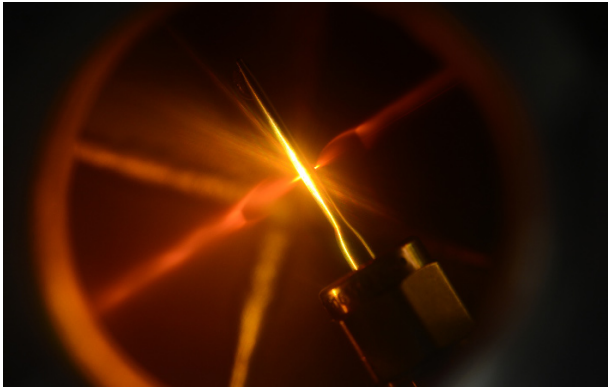


Figure 0.1: Generation of attosecond pulses at LAP (Thorsten Naeser, MPQ)

The Laboratory of Attosecond Physics (LAP) is a unique facility for research on ultrafast particle motions outside of the atomic core. LAP is a joint facility of the Max Planck Institute of Quantum Optics (MPQ) and the Ludwig-Maximilians-Universität (LMU) Munich. The LAP team includes 150 scientists and students. Many of them are organized within the DFG cluster of excellence Munich-Centre for Advanced Photonics (MAP). The scientists are mainly interested in the motion of electrons, which change their probability density in quantum mechanical steps within attoseconds. In order to record such motion, the physicists have developed light flashes that last only

attoseconds in duration. One attosecond is a billionth of a billionth of a second.

Physicists led by Prof. Dr. Ferenc Krausz, the current leader of the LAP team, generated and measured such extremely short light flashes for the first time in 2001. Since then, impressive insight into the mostly unknown world of electron motion has been gained world-wide, where the real-time dynamics of these particles can be followed after light-induced excitation.

The LAP research group Ultrafast Nanophotonics, which also Johannes Schötz is part of, is led by Prof. Dr. Matthias Kling. The research group investigates how electrons in complex materials collectively behave under the influence of intense laser light. In particular the physicists are interested in the dynamics and the control of electrons in molecules and nanostructures. Such light-induced electron motion occurs, for example, in semiconductors and dielectrics within attoseconds. Technically, the research could advance light-controlled nanoelectronics. With light frequencies in the Petahertz regime (10^{15} Hz) ultrafast switching times of electronic circuits could be achieved. This would advance current electronics by many (up to about 5) orders of magnitude.

Thorsten Naeser

Internet:

www.attoworld.de

www.munich-photonics.de

Contents

Introduction	1
2 Theoretical background	5
2.1 Ultrashort Laserpulses	5
2.2 Maxwell's equations	6
2.3 Nanoplasmonics	8
2.4 Mie Theory	10
2.5 Attosecond streaking	13
2.5.1 Fundamentals of attosecond streaking	13
2.5.2 Attosecond streaking from solids	16
3 Experimental methods and setup	23
3.1 Generation of ultrashort laserpulses	23
3.2 CEP-stabilization	25
3.3 High-Harmonic Generation	28
3.4 AS5-Beamline	31
4 Electron scattering in solids	35
4.1 Elastic Scattering	35
4.2 Inelastic Scattering	41
4.2.1 Kinematics of inelastic scattering	41
4.2.2 Theory	43
4.2.3 Extension algorithms	46
4.2.4 Energy Loss Function	49
4.3 Surface Scattering	53
4.4 Transmission	57
4.5 Simulation for a plane surface	59

5	Attosecond streaking from metal nanotips	63
5.1	General characteristics of nanoplasmonic streaking . .	63
5.2	Theoretical Modelling	66
5.3	Experiments	75
5.4	Analysis	77
5.5	Suggestion for proof-of-principle experiment	88
6	Conclusion and Outlook	91
	Appendix A: Description of electron scattering	93

List of Figures

1.1	CPU clock speeds	2
2.1	Few-cycle laser pulses	6
2.2	Surface plasmons	9
2.3	Principle of attosecond streaking	14
2.4	Experimental attosecond streaking trace	16
2.5	Three step picture of photoemission	17
2.6	Density of states of Au	20
2.7	Relation between DOS and photoemission spectra	21
3.1	Overview of the laser system	24
3.2	Carrier-envelope phase stabilization	26
3.3	Principle of high-harmonic generation	29
3.4	Isolated attosecond pulses from high-harmonic generation	31
3.5	Experimental XUV pulses	32
3.6	Attosecond beamline	32
3.7	Delay stage and TOF detector	33
4.1	Differential cross sections of elastic scattering	38
4.2	Higher order effects on elastic scattering	38
4.3	Dependence of elastic scattering on the muffin-tin radius	40
4.4	The kinematics of inelastic scattering.	42
4.5	Bulk loss function for gold	50
4.6	Mermin extension algorithm	51
4.7	Mean free path and differential cross section for inelastic scattering	51
4.8	Inelastic scattering: IMFP and DCS	52
4.9	Surface loss function	54

4.10	DSEP and SEP of inelastic surface scattering	56
4.11	Transmission through surface potential step	57
4.12	Simulation of surface scattering	59
4.13	Simulation results	61
5.1	Near-field decay	64
5.2	Near-field streaking regimes: phase and amplitude shift	67
5.3	Geometry of the nanotips	68
5.4	Electric near-fields around a nanotip and effect on attosecond streaking traces	70
5.5	linear electromagnetic response of the nanotip	72
5.6	Attosecond streaking simulation	73
5.7	Effect of geometry on the nanotip	76
5.8	Electron spectra from nanotips	78
5.9	Analysis of gas streaking traces	79
5.10	Attosecond streaking spectrum from a gold nanotip . .	80
5.11	Comparison of gas and nanotip streaking traces	81
5.12	Extracted peak shifts	82
5.13	Correlation between peak shifts and the IR period . .	83
5.14	Relative streaking amplitude	86
5.15	Different streaking geometries	88
5.16	Relative phase shift and amplitude in alternative ge- ometry	90