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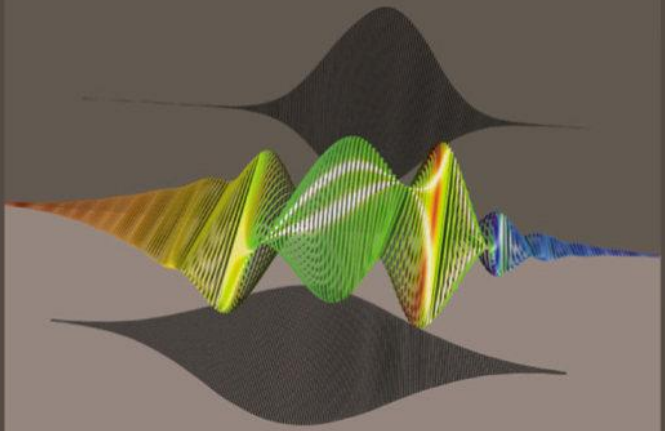

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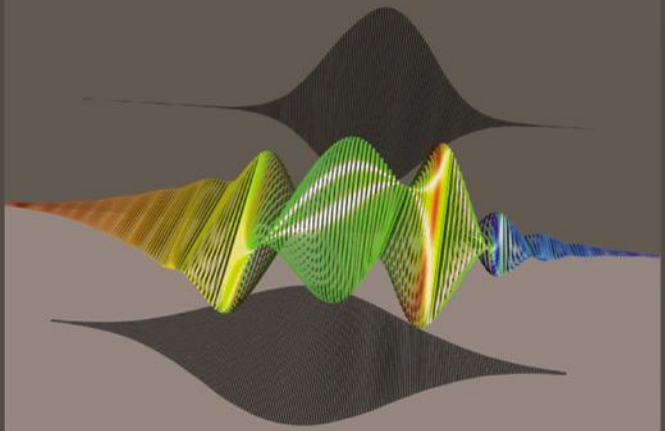
FEMTOSECOND LASER SPECTROSCOPY



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PETER HANNAFORD

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Femtosecond Laser Spectroscopy

Edited by
Peter Hannaford

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Contents

Contributing Authors

Number in parentheses indicates first page of author's contribution.

S. AKTURK (61), School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430, USA

J. B. ASBURY (167), Department of Chemistry, Stanford University, Stanford, CA 94305, USA

S.N. BAGAYEV (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

T. BAUMERT (225), Institute of Physics, University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

M. BELLINI (29), Istituto Nazionale di Ottica Applicata (INOA), Largo Fermi 6, 50125, Florence, Italy

T. BRIXNER (225), Physics Department, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

P. CANCIO (109), Istituto Nazionale di Ottica Applicata (INOA), Largo Fermi 6, 50125 Florence, Italy, and European Laboratory for Nonlinear Spectroscopy (LENS), Via Carrara 1, 50019 Sesto Fiorentino FI, Italy

Q. CAO (61), School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430, USA

B. CHATEL (267), Laboratoire de Collisions, Agrégats et Réactivité (CNRS UMR 5589), IRSAMC, Université Paul Sabatier, 31062 Toulouse CEDEX, France

L.V. DAO (197), Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, PO Box 218, Hawthorn, Victoria 3122, Australia

P. DE NATALE (109), Istituto Nazionale di Ottica Applicata (INOA), Largo Fermi 6, 50125 Florence, Italy, and European Laboratory for Nonlinear Spectroscopy (LENS), Via Carrara 1, 50019 Sesto Fiorentino FI, Italy

V.I. DENISOV (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

M.D. FAYER (167), Department of Chemistry, Stanford University, Stanford, CA 94305, USA

G. GERBER (225), Physics Department, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

B. GIRARD (267), Laboratoire de Collisions, Agrégats et Réactivité (CNRS UMR 5589), IRSAMC, Université Paul Sabatier, 31062 Toulouse CEDEX, France

X. GU (61), School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430, USA

P. HANNAFORD (197), Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, PO Box 218, Hawthorn, Victoria 3122, Australia

V.M. KLEMENTYEV (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

T. KOBAYASHI (133), Department of Physics, University of Tokyo, Hongo 7-3-1, Bunkyo, Tokyo 113-0033, Japan

I.I. KOREL (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

S.A. KUZNETSOV (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

C. LINCOLN (197), Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, PO Box 218, Hawthorn, Victoria 3122, Australia

M. LOWE (197), Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, PO Box 218, Hawthorn, Victoria 3122, Australia

D. MAZZOTTI (109), Istituto Nazionale di Ottica Applicata (INOA), Largo Fermi 6, 50125 Florence, Italy, and European Laboratory for Nonlinear Spectroscopy (LENS), Via Carrara 1, 50019 Sesto Fiorentino FI, Italy

M. MERO (305), Department of Physics and Astronomy, University of New Mexico, Albuquerque NM 87131, USA

T. PFEIFER (225), Physics Department, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

V.S. PIVTSOV (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

W. RUDOLPH (305), Department of Physics and Astronomy, University of New Mexico, Albuquerque NM 87131, USA

A. SHREENATH (61), School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430, USA

T. STEINEL (167), Department of Chemistry, Stanford University, Stanford, CA 94305, USA

R. TREBINO (61), School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430, USA

M. WOLLENHAUPT (225), Institute of Physics, University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

J. YE (1), JILA, National Institute of Standards and Technology and University of Colorado, Boulder, Colorado 80309-0440, USA

V.F. ZAKHARYASH (87), Institute of Laser Physics, Siberian Branch, Russian Academy of Sciences, Pr. Lavrentieva, 13/3, 630090 Novosibirsk, Russia

J. ZELLER (305), Department of Physics and Astronomy, University of New Mexico, Albuquerque NM 87131, USA

Foreword

The embryonic development of femtoscience stems from advances made in the generation of ultrashort laser pulses. Beginning with mode-locking of glass lasers in the 1960s, the development of dye lasers brought the pulse width down from picoseconds to femtoseconds. The breakthrough in solid state laser pulse generation provided the current reliable table-top laser systems capable of average power of about 1 watt, and peak power density of easily $10^{12} - 10^{13}$ watts per square centimeter, with pulse widths in the range of four to eight femtoseconds. Pulses with peak power density reaching 10^{20} watts per square centimeter have been achieved in laboratory settings and, more recently, pulses of sub-femtosecond duration have been successfully generated.

As concepts and methodologies have evolved over the past two decades, the realm of ultrafast science has become vast and exciting and has impacted many areas of chemistry, biology and physics, and other fields such as materials science, electrical engineering, and optical communication. In molecular science the explosive growth of this research is for fundamental reasons. In femtochemistry and femtobiology chemical bonds form and break on the femtosecond time scale, and on this scale of time we can freeze the transition states at configurations never before seen. Even for non-reactive physical changes one is observing the most elementary of molecular processes. On a time scale shorter than the vibrational and rotational periods the ensemble behaves coherently as a single-molecule trajectory.

But these developments would not have been possible without the crystallization of some key underlying concepts that were in the beginning shrouded in fog. First was the issue of the “uncertainty principle”, which had to be decisively clarified. Second was the question of whether one could

sustain wave-packet motion at the atomic scale of distance. In other words, would the de Broglie wavelength of the atom become sufficiently short to define classical motion – "classical atoms" – and without significant quantum spreading? This too had to be clearly demonstrated and monitored in the course of change, not only for elementary processes in molecular systems, but also during complex biological transformations. And, finally, some questions about the uniqueness and generality of the approach had to be addressed. For example, why not deduce the information from high-resolution frequency-domain methods and then Fourier transform to obtain the dynamics? It is surely now clear that transient species cannot be isolated this way, and that there is no substitute for direct "real time" observations that fully exploit the intrinsic coherence of atomic and molecular motions.

Theory has enjoyed a similar explosion in areas dealing with *ab initio* electronic structures, molecular dynamics, and nonlinear spectroscopies. There has been progress in calculating potential energy surfaces of reactive systems, especially in their ground state. On excited-state surfaces it is now feasible to map out regions of the surface where transition states and conical intersections are important for the outcome of change. For dynamics, new methods have been devised for direct viewing of the motion by formulating the time-dependent picture, rather than solving the time-independent Schrödinger equation and subsequently constructing a temporal picture. Analytical theory has been advanced, using time-ordered density matrices, to enable the design of multidimensional spectroscopy, the analogue of 2-D (and higher) NMR spectroscopy. That the coupling between theory and experiment is profound is evident in many of the chapters in this volume.

Other areas of studies are highlighted in this volume. The making of femtosecond combs for precision metrology and spectroscopy, and the advances in nonlinear and multidimensional optical techniques are two examples of such frontiers. The ability to count optical oscillations of more than 10^{15} cycles per second can potentially provide all-optical atomic clocks with a new limit of precision. Similarly, the ability to generate sub-femtosecond pulses pushes the limit and resolution toward new studies of electron dynamics. Besides these advances in precision (optical cycles) and pulse duration (pulse width) there are those concerned with the phase. Beginning in 1980, the phase of an optical pulse has been experimentally under control and pulses of well-defined phases (π, $\pi/2$, etc) have been generated and utilized in, among other applications, the control of emission from molecules. But only recently could composite phases be prescribed with a feedback algorithm to control the outcome of a reactive channel, as shown in this volume. Coherent control is a frontier field stimulating research in both theory and experiment.

Edited by Peter Hannaford this volume is a welcomed edition to the field as it brings together the latest in some areas of developments with an impressive mix of new methodologies and applications. The use of femtosecond combs for precision measurements is well covered and coherent control is presented with demonstrations for atomic, molecular and electronic processes. Nonlinear optical methods, including novel geometries of photon and vibrational echoes, are described for the investigation of molecular systems, in particular dye molecules, hydrogen-bonded networks, semiconductor quantum dots, and biomolecules. Measurements of ultrashort pulses, time-resolved reflection and transmission methods, and real-time spectroscopy with sub-5-femtosecond visible pulses provide the means for exploring new regimes and resolutions.

This book in the series on *Progress in Lasers* gives an exposé of some current and exciting research areas in the technology of pulse generation and in the applications of femtoscience.

Ahmed Zewail
California Institute of Technology
Pasadena, California
May 2004

Preface

When I was first approached to edit a volume on *Femtosecond Laser Spectroscopy* in 2000, I did not anticipate that the field was about to explode, with the announcement of a series of remarkable new developments and advances. This volume describes these recent developments in eleven chapters written by leading international researchers in the field. It includes sections on:

- Femtosecond optical frequency combs, which are currently revolutionising ultrahigh precision spectroscopy and optical frequency metrology;
- Soft X-ray femtosecond laser sources, which promise to have important applications in biomedical imaging;
- Attosecond laser sources, which will provide the next generation of sources to study ultrafast phenomena such as electron dynamics;
- Novel methods for measuring and characterizing ultrashort laser pulses and ultrashort pulses of light;
- Coherent control of atomic, molecular and electron dynamics with tailored femtosecond laser pulses;
- Real-time Spectroscopy of molecular vibrations with sub-5-fs pulses; and
- Multidimensional femtosecond coherent spectroscopies for studying molecular and electron dynamics.

Indeed, it is gratifying to see that with the recent advent of attosecond laser sources the title of this volume may soon be rendered obsolete.

I would like to thank each of the contributors for their cooperation in preparing this volume, and Ahmed Zewail for writing the Foreword. I appreciate the amount of work that goes into writing chapters of this type when the authors are heavily burdened with other demands on their time. I

feel honoured and privileged to have been associated with such an eminent group of researchers. I also thank my co-workers in the Ultrafast Spectroscopy group at Swinburne University of Technology – Lap Van Dao, Martin Lowe, Craig Lincoln, Shannon Whitlock, Xiaoming Wen, Tra My Do, Petrissa Eckle and David McDonald – for their help and encouragement during the preparation of this volume and for critical reading of some of the chapters. I thank Tien Kieu, Grainne Duffy and David Lau for their assistance with the preparation of the camera-ready chapters, and Gustav Gerber for kindly allowing the use of Figure 9-12 on the front cover of this volume. Finally, I thank the publishers of the following journals and books for permission to reproduce material in this volume: Applied Physics B, Applied Physics Letters, Journal of Chemical Physics, Journal of Physics B, Laser Spectroscopy Proceedings, Nature, Optics Express, Optics Letters, Optical Review, Review of Scientific Instruments, Physical Review Letters, Physical Review A, and SPIE Proceedings.

Peter Hannaford
Swinburne University of Technology
Melbourne, June 2004

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