Ultrafast Phenomena VIII

Proceedings of the 8th International Conference, Antibes Juan-Les-Pins, France, June 8–12, 1992

With 523 Figures

Springer-Verlag
Berlin Heidelberg New York
London Paris Tokyo
Hong Kong Barcelona
Budapest
## Contents

### Part I  Overview and General Prospects

<table>
<thead>
<tr>
<th>Title</th>
<th>Author(s)</th>
<th>Pages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Additive Pulse Modelocking and Kerr-Lens Modelocking</td>
<td>H.A. Haus (With 6 Figures)</td>
<td>3</td>
</tr>
<tr>
<td>Molecular Control Spectrometer</td>
<td>Y. Yan, B.E. Kohler, R.E. Gillilan, R.M. Whitnell, K.R. Wilson, and S. Mukamel</td>
<td>8</td>
</tr>
<tr>
<td>Internal Motions of Proteins</td>
<td>M. Karplus</td>
<td>13</td>
</tr>
<tr>
<td>Some Theoretical Aspects of Electron Transfer in Supermolecules</td>
<td>J. Jortner and M. Bixon (With 3 Figures)</td>
<td>15</td>
</tr>
<tr>
<td>Femtosecond Time-Resolved Spectroscopy of Magneto-Excitons</td>
<td>D.S. Chemla, J.B. Stark, and W.H. Knox (With 6 Figures)</td>
<td>21</td>
</tr>
<tr>
<td>High-Order Harmonic Generation in Strong Laser Fields</td>
<td>A. L'Huillier and P. Balcou (With 3 Figures)</td>
<td>29</td>
</tr>
<tr>
<td>QED at $10^{20}$ W/cm²</td>
<td>A.C. Melissinos (With 6 Figures)</td>
<td>34</td>
</tr>
</tbody>
</table>

### Part II  Elementary Dynamics: Chemistry, Biology and Physics

<table>
<thead>
<tr>
<th>Title</th>
<th>Author(s)</th>
<th>Pages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Femtochemistry</td>
<td>A.H. Zewail (With 6 Figures)</td>
<td>43</td>
</tr>
</tbody>
</table>
Mechanisms of Charge Separation in Bacterial Reaction Centers
By M.H. Vos, F. Rappaport, J.-C. Lambry, C. Rischel, J. Breton, and J.-L. Martin (With 2 Figures) .......................... 58

Coherent Phonons in Superconducting Materials
By W. Albrecht, Th. Kruse, and H. Kurz (With 3 Figures) ............. 63

Displacive Excitation of Coherent Phonons
By T.K. Cheng, J. Vidal, H.J. Zeiger, E.P. Ippen, G. Dresselhaus, and M.S. Dresselhaus (With 1 Figure) ............... 66

Femtosecond Time-Resolved Photodissociation of Triiodide Ions in Alcohol Solution: Directly Observed Photoinduced Vibrational Coherence of Reactants and Products
By U. Banin, A. Waldman, and S. Ruhman (With 4 Figures) .......... 68

Vibrational Coherence in Charge Transfer
By K. Wynne, C. Galli, P.J.F. De Rege, M.J. Therien, and R.M. Hochstrasser (With 1 Figure) ......................... 71

Ultrafast Dynamics in Solution:
Wavepacket Motion and the Cage Effect in Iodine
By Y. Yan, R.M. Whitnell, K.R. Wilson, and A.H. Zewail
(With 1 Figure) ................................................... 74

Femtosecond Time-Resolved Ionization Spectroscopy of Polyatomic Molecules
By M. Seel and W. Domcke (With 1 Figure) .......................... 76

A Study of Nuclear Vibrational Wave Packets in Na₂
by Time- and Frequency-Resolved Fluorescence Upconversion
By I.A. Walmsley, T.J. Dunn, J. Sweetser, and C. Radzewicz
(With 3 Figures) .................................................. 78

Ultrafast Dynamics of Solid C₆₀
By S.L. Dexheimer, D.M. Mittleman, R.W. Schoenlein, W. Vareka, X.-D. Xiang, A. Zettl, and C.V. Shank (With 2 Figures) ............. 81

Femtosecond Dynamics
of Molecular and Cluster Ionization and Fragmentation
By T. Baumert, R. Thalweiser, V. Weiβ, and G. Gerber
(With 5 Figures) .................................................. 83

Dephasing and Beats of Excitonic-Enhanced Transitions of J-Aggregates Measured by Femtosecond Time-Resolved Resonance CARS
By V.F. Kamalov, R. Inaba, and K. Yoshihara (With 1 Figure) .......... 87

Excited States Dynamics of the Special Pair Dimer
By P.O.J. Scherer and S.F. Fischer (With 4 Figures) ................. 89

Creation of an Anti-Wavepacket in a Rydberg Atom
(With 3 Figures) .................................................. 92

VIII
Squeezing of the Molecular Vibrations by Femtosecond Laser Pulses
By A.V. Vinogradov and J. Janszky (With 1 Figure) .............................. 95

Part III Spectroscopy and Advances in Measurements

Spectroscopic Applications of Phase-Locked Femtosecond Pulses
By N.F. Scherer, M. Cho, L.D. Ziegler, M. Du, A. Matro, J. Cina,
and G.R. Fleming (With 5 Figures) .................................................. 99

Use of Piecewise Phase-Swept Pulses
to Counteract Inhomogeneous Decay in Wave Packet Interferometry
By L.W. Ungar, A. Matro, and J.A. Cina (With 1 Figure) ..................... 105

Ultrafast Nonlinear Spectroscopy with Chirped Optical Pulses
By E.T.J. Nibbering, F. de Haan, D.A. Wiersma, and K. Duppen
(With 2 Figures) ............................................................................... 107

Multiple Excitation Pulse,
Multiple Probe Pulse Femtosecond Spectroscopy
By G.P. Wiederrecht, W. Wang, K.A. Nelson, A.M. Weiner,
and D.E. Leaird (With 2 Figures) ....................................................... 110

Stimulated Emission Pumping and Selective Excitation by Adiabatic
Passage with Frequency-Modulated Picosecond Laser Pulses
By J.S. Melinger, A. Hariharan, S.R. Gandhi, and W.S. Warren
(With 2 Figures) ............................................................................... 113

A Subpicosecond Optical Sampling System
By J.D. Kafka, J.W. Pieterse, and M.L. Watts (With 2 Figures) ............... 116

Femtosecond Sagnac Interferometry
By J.-C. Diels, P. Dorn, M. Lai, W. Rudolph, and X.M. Zhao
(With 3 Figures) ............................................................................... 120

Femtosecond Time-Gated Imaging of Translucent Objects
Hidden in Highly Scattering Media
By K.M. Yoo, B.B. Das, F. Liu, Q. Xing, and R.R. Alfano
(With 2 Figures) ............................................................................... 124

Femtosecond Waveform Processing via Spectral Holography
(With 4 Figures) ............................................................................... 128

The Chronocyclic Representation of Ultrashort Light Pulses
By J. Paye (With 4 Figures) .............................................................. 133

Femtosecond Pulse Phase Measurement
by Spectrally Resolved Up-Conversion
By J.-P. Foing, J.-P. Likforman, and M. Joffre (With 3 Figures) .............................. 136
<table>
<thead>
<tr>
<th>Title</th>
<th>Authors</th>
<th>Pages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single-Shot Measurement of the Intensity and Phase of a Femtosecond Pulse</td>
<td>D.J. Kane and R. Trebino</td>
<td>138</td>
</tr>
<tr>
<td>Two-Photon Interference Measurement of Ultrafast Laser Pulses</td>
<td>M. Matsuoka, Y. Miyamoto, T. Kuga, M. Baba, and Y. Li</td>
<td>140</td>
</tr>
<tr>
<td>Picosecond Single-Shot Pulse-Shape Measurement by Stochastic Sampling of Detected Photon Times</td>
<td>N. Adams, C. Bovet, E. Rossa, and A. Simonin</td>
<td>142</td>
</tr>
<tr>
<td>Integrated Devices for Single Picosecond Pulse Measurements</td>
<td>V. Gerbe, M. Cuzin, M.C. Gentet, and J. Lajzerowicz</td>
<td>145</td>
</tr>
<tr>
<td>Distortion of a 6 fs Pulse in the Focus of a BK7 Lens</td>
<td>Zs. Bor and Z.L. Horváth</td>
<td>150</td>
</tr>
</tbody>
</table>

**Part IV  Tools: Sources and Amplifiers**

<table>
<thead>
<tr>
<th>Title</th>
<th>Authors</th>
<th>Pages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modelocking, Stabilizing, and Starting Ultrashort Pulse Lasers</td>
<td>E.P. Ippen</td>
<td>155</td>
</tr>
<tr>
<td>Design Considerations for Femtosecond Ti:Sapphire Oscillators</td>
<td>Ch. Spielmann, P.F. Curley, T. Brabec, E. Wintner, A.J. Schmidt, and F. Krausz</td>
<td>163</td>
</tr>
<tr>
<td>Self-Mode-Locked Cr³⁺:LiCaAlF₆ and Cr³⁺:LiSrAlF₆ Lasers</td>
<td>A. Miller, P. Li Kam Wa, B.H.T. Chai, J.M. Evans, and W. Sibbett</td>
<td>166</td>
</tr>
<tr>
<td>Sub-50 fs Pulse Generation from a Self-Starting CW Passively Mode-Locked Cr:LiSrAlF₆ Laser</td>
<td>N.H. Rizvi, P.M.W. French, and J.R. Taylor</td>
<td>169</td>
</tr>
<tr>
<td>CW Krypton-Laser Pumped Cr³⁺:LiSrAlF₆ and Cr³⁺:LiSr₀.₈Ca₀.₂AlF₆ Crystals Produce 150 fs Mode-Locked Pulses</td>
<td>A. Miller, P. Li Kam Wa, H.S. Wang, S.L. Ayres, E.W. Van Stryland, and B.H.T. Chai</td>
<td>172</td>
</tr>
</tbody>
</table>
60-fs Chromium-Doped Forsterite (Cr$^{4+}$:Mg$_2$SiO$_4$) Laser
By A. Seas, V. Petričević, and R.R. Alfano (With 3 Figures) ............ 174

Femtosecond Pulses from Nd:Glass Lasers
By A.J. Schmidt, M.H. Ober, M. Hofer, M.E. Fermann, F. Krausz,
T. Brabec, Ch. Spielmann, and E. Wintner (With 3 Figures) ............ 177

A Diode-Pumped Picosecond Oscillator at 1053 nm
By I.P. Mercer, Z. Chang, M.R.G. Miller, C.N. Danson, C.B. Edwards,
and M.H.R. Hutchinson (With 3 Figures) ............................. 182

A New Intracavity Antiresonant Semiconductor Fabry-Perot Passively Mode-Locks Nd:YLF and Nd:YAG Lasers
By U. Keller, D.A.B. Miller, G.D. Boyd, T.H. Chiu, J.F. Ferguson,
and M.T. Asom (With 3 Figures) ........................................... 184

CW Mode-Locked Singly-Resonant Optical Parametric Oscillator Pumped by a Ti:Sapphire Laser
By A. Nebel, U. Socha, and R. Beigang (With 1 Figure) ................. 187

70 fs, High-Average Power, CW Infrared Optical Parametric Oscillator
By G. Mak, Q. Fu, and H.M. van Driel (With 2 Figures) .................. 190

Femtosecond Intracavity Dispersion Measurements
By W.H. Knox (With 2 Figures) ............................................. 192

Time Synchronization Measurements
Between Two Self-Modelocked Ti:Sapphire Lasers
By D.E. Spence, W.E. Sleat, J.M. Evans, W. Sibbett, and J.D. Kafka
(With 2 Figures) ............................................................... 194

Femtosecond Synchronous Pumping of Dye Lasers with <100 fs Jitter
By W.H. Knox and F.A. Beisser (With 2 Figures) .......................... 196

Development of High Average Power Femtosecond Amplifiers Based on Ti:, Cr: and Nd:Doped Materials
By J. Squier, S. Coe, G. Mourou, D. Harter, and F. Salin ............... 198

Femtosecond Pulse Amplification and Continuum Generation at >250 kHz with a Ti:Sapphire Regenerative Amplifier
By T.B. Norris (With 4 Figures) .............................................. 200

Millijoule Femtosecond Pulse Amplification in Ti:Al$_2$O$_3$
at Multi-kHz Repetition Rates
By F. Salin, J. Squier, G. Mourou, and G. Vaillancourt
(With 4 Figures) ............................................................... 203

High Repetition Rate CW Pumped Cr:LiSAF Regenerative Amplifier
By F. Balembois, P. Georges, F. Salin, G. Roger, and A. Brun
(With 4 Figures) ............................................................... 206
18 fs Pulse Generation
by a Single Excimer-Laser-Pumped Pulsed Dye Laser
By P. Simon, C. Jordan, and S. Szatmari (With 2 Figures) 209

Monolithic CPM Diode Lasers
By M.C. Wu, Y.K. Chen, T. Tanbun-Ek, and R.A. Logan
(With 5 Figures) 211

Ultrashort Pulse Generation from High-Power Arrays
Using Intracavity Nonlinearities
By L.Y. Pang, J.G. Fujimoto, and E.S. Kintzer (With 3 Figures) 217

100-Gbps Response of Microcavity Lasers
By H. Yokoyama, Y. Nambu, and T. Shimizu (With 2 Figures) 220

Sequential Laser Emission
in Multiple Quantum Well Vertical-Cavity Structures
By C. Tanguy, J.-L. Oudar, B. Sermage, and R. Azoulay
(With 2 Figures) 222

Experimental Analysis of Gain Modulation
in Sub-Picosecond (~0.45 ps) Mode-Locked Laser Diodes
By N. Stelmakh, J.-M. Lourtioz, and D. Pascal (With 3 Figures) 224

Generation of Stable Pulse Trains
with a Passively Modelocked Er-Fiber Laser
By M.E. Fermann, M.J. Andrejco, Y. Silberberg, and A.M. Weiner
(With 4 Figures) 227

Generation of Pairs of Solitons
in an All-Fibre, Femtosecond Soliton Source
By D.J. Richardson, V.V. Afanasjev, A.B. Grudinin, and D.N. Payne
(With 5 Figures) 229

Nonlinear Loop Mirrors in Fiber Lasers
(With 4 Figures) 232

Temporal Characteristics of the Ytterbium–Erbium Figure-8 Laser
By I.Yu. Khrushchev, A.B. Grudinin, and E.M. Dianov
(With 3 Figures) 235

Generation of 1.7 ps Solitons by Amplification of Pulses from a Laser
Diode with Saturable Absorber in an Erbium-Doped Fibre

Part V  High Intensity and Nonlinear Effects

Generation of Ultra-Intense Pulses and Applications
By G. Mourou (With 1 Figure) 241
Generation of 50 TW Femtosecond Pulses in a Nd-Glass Chain
By C. Rouyer, E. Mazataud, I. Allais, A. Pierre, and S. Seznec
(With 2 Figures) .................................................. 248

All-Solid Femtosecond Oscillator-Amplifier Laser Chain
with 100 mJ per Pulse
By C. Le Blanc, G. Grillon, J.P. Chambaret, G. Boyer, M. Franco,
A. Mysyrowicz, and A. Antonetti (With 1 Figure) .................. 251

Development of a High Intensity Femtosecond LiSAF Laser
By M.C. Richardson, P. Beaud, B.H.T. Chai, E. Miesak, Y.-F. Chen,
and V. Yanovsky (With 2 Figures) ................................. 253

Contrasted Behaviors of Stark-Induced Resonances
in Multiphoton Ionization of Krypton
By E. Mevel, R. Trainham, J. Breger, G. Petite, P. Agostini,
J.P. Chambaret, A. Migus, and A. Antonetti (With 1 Figure) .... 255

Phase-Dependent Ionization Using an Intense Two-Color Light Field
By D. Schumacher, M.P. de Boer, H.G. Muller, R.R. Jones,
and P.H. Bucksbaum (With 2 Figures) .............................. 257

Stabilization of Atoms in Ultra-Intense Laser Pulses: A Classical Model
By A. Maquet, T. Ménis, R. Taïeb, and V. Véniard (With 1 Figure) ... 259

Inertially Confined Molecular Ions
By M. Laberge, P. Dietrich, and P.B. Corkum (With 2 Figures) .... 261

A Femtosecond Lightning Rod
By X.M. Zhao, C.Y. Yeh, J.-C. Diels, and C.Y. Wang
(With 2 Figures) ..................................................... 264

Plasma Physics with Ultra-Short and Ultra-Intense Laser Pulses
By T.W. Johnston, Y. Beaudoin, M. Chaker, C.Y. Côté, J.C. Kieffer,
J.P. Matte, H. Pépin, C.Y. Chien, S. Coe, G. Mourou, and D. Umstadter
(With 1 Figure) ..................................................... 267

X-Rays Generated by Femtosecond Laser-Produced Plasmas
By J.P. Geindre, P. P. Audebert, A. Rousse, F. Fallières, J.C. Gauthier,
A. Mysyrowicz, G. Grillon, J.P. Chambaret, A. Antonetti, A. Mens,
R. Verrecchia, R. Sauneuf, and P. Schirman (With 2 Figures) ...... 272

K-Shell Emission from 100 fs Laser-Produced Plasmas
Created from Porous Aluminum Targets
By R. Shepherd, D. Price, B. White, S. Gordan, A. Osterheld, R. Walling,
D. Slaughter, and R. Stewart (With 2 Figures) ...................... 275

Kilovolt X-Ray Emission from Femtosecond Laser-Produced Plasmas
By G. Jenke, H. Schüler, T. Engers, D. von der Linde, I. Uschmann,
E. Förster, and K. Gäbel (With 1 Figure) ......................... 278
<table>
<thead>
<tr>
<th>Title</th>
<th>Authors</th>
<th>Pages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photon Acceleration via Laser-Produced Ionization Fronts</td>
<td>By R.L. Savage Jr., R.P. Brogle, W.B. Mori, and C. Joshi</td>
<td>286</td>
</tr>
<tr>
<td>Propagation of Intense Laser Pulses in Plasmas</td>
<td>By E. Esarey, P. Sprangle, J. Krall, and G. Joyce (With 1 Figure)</td>
<td>290</td>
</tr>
<tr>
<td>Ponderomotive Steepening in Short-Scale-Length Laser-Plasmas</td>
<td>By D. Umstadter and X. Liu (With 2 Figures)</td>
<td>293</td>
</tr>
<tr>
<td>Possibility of Experimental Studies of Nonlinear Quantum Electrodynamics Effects Using High Power Ultrashort Laser Pulses</td>
<td>By P.G. Kryukov (With 1 Figure)</td>
<td>296</td>
</tr>
<tr>
<td>Soliton-Like Self-Trapping of Three-Dimensional Patterns</td>
<td>By A. Barthelemy, C. Froehly, M. Shalaby, P. Donnat, J. Paye, and A. Migus  (With 9 Figures)</td>
<td>299</td>
</tr>
<tr>
<td>Efficient Raman Conversion of Femtosecond UV Light Pulses</td>
<td>By K.A. Stankov and Y.-W. Lee (With 1 Figure)</td>
<td>311</td>
</tr>
<tr>
<td>Organic Crystalline Fiber for Efficient Compression of Femtosecond Laser Pulses</td>
<td>By M. Yamashita (With 1 Figure)</td>
<td>313</td>
</tr>
<tr>
<td>Nonlinear Temporal Diffraction in Optical Fibers</td>
<td>By G.R. Boyer, M.K. Jackson, J. Paye, M.A. Franco, and A. Mysyrowicz (With 3 Figures)</td>
<td>315</td>
</tr>
<tr>
<td>Title</td>
<td>Authors</td>
<td>Pages</td>
</tr>
<tr>
<td>----------------------------------------------------------------------</td>
<td>------------------------------------------------------------------------</td>
<td>-------</td>
</tr>
<tr>
<td>Experimental Investigation of Dark Solitons Interaction</td>
<td>Ph. Emplit, J.-P. Hamaide, and M. Haelterman (With 3 Figures)</td>
<td>320</td>
</tr>
<tr>
<td>Compression of Pulses from Soliton Fibre Lasers in a Dispersion-Decreasing Fibre</td>
<td>S.V. Chernikov, D.J. Richardson, E.M. Dianov, and D.N. Payne (With 4 Figures)</td>
<td>325</td>
</tr>
<tr>
<td>Observation of the Thermalization of Electrons in a Metal Excited by Femtosecond Optical Pulses</td>
<td>W.S. Fann, R. Storz, H.W.K. Tom, and J. Bokor (With 2 Figures)</td>
<td>331</td>
</tr>
<tr>
<td>Electron–Electron Dynamics Observed in Femtosecond Thermoreflection Measurements on Noble Metals</td>
<td>R.H.M. Groeneveld, R. Sprik, and Ad. Lagendijk (With 2 Figures)</td>
<td>338</td>
</tr>
<tr>
<td>Inversion of Single- and Two-Photon Photoelectric Sensitivities of Metals in the Femtosecond Range</td>
<td>J.P. Girardeau-Montaut, C. Girardeau-Montaut, S.D. Moustaïzis, and C. Fotakis (With 1 Figure)</td>
<td>340</td>
</tr>
<tr>
<td>Femtosecond Relaxation of Plasma Excitations in Silver Films</td>
<td>R.A. Höpfel, D. Steinmüller-Nethl, F.R. Aussennegg, and A. Leitner (With 3 Figures)</td>
<td>342</td>
</tr>
<tr>
<td>Femtosecond Free Induction Decay of Metal Surface Adsorbate Vibrations</td>
<td>J.C. Owrutsky, J.P. Culver, M. Li, Y.R. Kim, M.J. Sarisky, M.S. Yeganeh, R.M. Hochstrasser, and A.G. Yodh (With 1 Figure)</td>
<td>345</td>
</tr>
<tr>
<td>Observation of Laser-Induced Desorption of CO from Cu(111) with 100 fs Time-Resolution</td>
<td>J.A. Prybyla, H.W.K. Tom, and G.D. Aumiller (With 2 Figures)</td>
<td>347</td>
</tr>
<tr>
<td>Femtosecond Desorption of Molecular Oxygen from Pt(111)</td>
<td>F.-J. Kao, D.G. Busch, D. Gomes da Costa, D. Cohen, and W. Ho (With 1 Figure)</td>
<td>350</td>
</tr>
</tbody>
</table>

Part VI  Metals, Surfaces and Materials
Femtosecond Carrier Dynamics in Solid C_{60} Films
By S.D. Brorson, M.K. Kelly, U. Wenschuh, R. Buhleier, and J. Kuhl
(With 4 Figures) 354

The Role of Covalency in Femtosecond Time-Resolved Reflectivity of Hydrodynamically Expanding Solid Surfaces
By X.Y. Wang, H.Y. Ahn, and M.C. Downer (With 1 Figure) 357

Ultrafast Formation Processes of Self-Trapped Excitons in Alkali Iodide Crystals under Band-to-Band Excitation
By T. Tokizaki, S. Iwai, T. Shibata, A. Nakamura, K. Tanimura, and N. Itoh (With 2 Figures) 360

Femtosecond Self-Trapping of Interacting Electron–Hole Pairs in α-SiO_{2}

Ultrafast Soft Mode Dynamics in Ferroelectric Crystals
By G.P. Wiederrecht, T.P. Dougherty, and K.A. Nelson (With 3 Figures) 365

Temporal Domain Study of the Phase Transition in PbTiO_{3}: A_1 Symmetry Investigation
By D.P. Kien, J.C. Loulergue, and J. Etchepare (With 2 Figures) 368

Femtosecond Transient Absorption Measurements on Low Band Gap Thiophene Polymers

Effects of Crosslinking in Host Polymer on Picosecond Optical Dephasing of Doped Dye Molecules
By S. Nakanishi, S. Fujiwara, M. Kawase, and H. Itoh (With 3 Figures) 372

Ultrafast Relaxation of Exciton and Soliton–Antisoliton Pair in One-Dimensional Conjugated Polymers
By T. Kobayashi, M. Yoshizawa, S. Takeuchi, and A. Yasuda (With 2 Figures) 376

Polarization-Dependent Femtosecond Dynamics of MBE-Grown Phthalocyanine Organic Thin Films
By Sandalphon, V.S. Williams, K. Meissner, N.R. Armstrong, and N. Peyghambarian (With 3 Figures) 379

Detection of a New Strongly-Coupled Vibration Mode During the Exciton Bleaching of Polydiacetylene
By J.M. Nunzi, C. Hirlimann, and J.F. Morhange (With 1 Figure) 381
Pressure-Induced Vibrational Relaxation and Electronic Dephasing in Molecular Crystals
By E.L. Chronister and R.A. Crowell (With 3 Figures) ....................... 384

Ultrafast Reversible Phase Changes for Optical Recording
By J. Solis, C.N. Afonso, F. Catalina, and C. Kalpouzos
(With 1 Figure) ............................ 387

Picosecond Transient Absorption and Fluorescence Emission Studies of C_{60} and C_{70} in Solution
By D. Kim, Y.D. Suh, S.K. Kim, and M. Lee (With 2 Figures) ........... 389

Part VII Semiconductors, Confinement and Opto-Electronics

Transient Absorption-Edge Singularities in GaAs
By D. Hulin, J.-P. Foing, M. Joffre, M.K. Jackson, J.-L. Oudar,
C. Tanguy, and M. Combescot (With 3 Figures) ......................... 395

Nonthermal Distribution of Electrons in GaAs
By D. Snoke and W.W. Rühle (With 1 Figure) ........................... 399

Femtosecond Carrier–Carrier Interaction in GaAs
By T. Gong, K.B. Ucer, L.X. Zheng, G.W. Wicks, J.F. Young, P.J. Kelly,
and P.M. Fauchet (With 4 Figures) ........................................ 402

Quantum Beats versus Polarization Interference:
An Experimental Distinction
By M. Koch, J. Feldmann, G. von Plessen, E.O. Göbel, P. Thomas,
and K. Köhler (With 1 Figure) ........................................... 405

Plasmon–Phonon Coupling and Hot Carrier Relaxation in GaAs and Low-Temperature-Grown GaAs
By R.I. Devlen, J. Kuhl, and K. Ploog (With 2 Figures) ................. 408

Femtosecond Carrier–Carrier Interaction Dynamics in Doped GaAs
By T. Furuta and A. Yoshii (With 1 Figure) ............................... 410

Femtosecond Carrier Kinetics in Low-Temperature-Grown GaAs
By X.Q. Zhou, H.M. van Driel, A.P. Heberle, W.W. Rühle, and K. Ploog
(With 2 Figures) ............................................................. 412

Transient Anisotropic Luminescence and Long-Living Polarization of an Optically Excited Dense Electron–Hole Plasma
By A.L. Ivanov and H. Haug (With 2 Figures) ............................... 414

Hot Hole Capture by Shallow Acceptors in p-Type GaAs
Studied by Picosecond Infrared Spectroscopy
By A. Lohner, M. Woerner, T. Elsaesser, and W. Kaiser
(With 2 Figures) ............................................................. 416
Ultrafast Dephasing and Interference of Coherent Phonons in GaAs
By W. Kütt, T. Pfeifer, T. Dekorsy, and H. Kurz (With 2 Figures) .... 418

Femtosecond, Electronically-Induced Disordering of GaAs
By J.-K. Wang, Y. Siegal, P.N. Saeta, N. Bloembergen, and E. Mazur
(With 2 Figures) ........................................... 420

Laser-Induced Ultrafast Order-Disorder Transitions in Semiconductors
By K. Sokolowski-Tinten, J. Bialkowski, and D. von der Linde
(With 1 Figure) ............................................. 422

Femtosecond Carrier Dynamics in InGaAsP Optical Amplifiers
By J. Mark and J. Mørk (With 1 Figure) .......................... 424

Ultrafast Nonlinear Refraction in Semiconductor Laser Amplifiers
By M. Sheik-Bahae and E.W. Van Stryland (With 3 Figures) ....... 426

Femtosecond Luminescence Spectroscopy of Indium Phosphide
By E. Fazio and G.M. Gale (With 2 Figures) .......................... 429

Dynamics of Excitons Probed by Accumulated Photon Echo
By T. Bouma, P. Vledder, and J.I. Dijkhuis (With 1 Figure) ....... 431

Time-Resolved Measurement of Hot Carrier Cooling Rates
in a-Si:H and a-Ge:H
By M. Wraback and J. Tauc (With 2 Figures) .......................... 433

Dephasing of the Short Exciton–Polariton Pulses
in Polar Semiconductors: The Cuprous Chloride Case
By F. Vallée, F. Bogani, and C. Flytzanis (With 3 Figures) ....... 435

Femtosecond Electronic Dynamics of CdSe Nanocrystals
By C.V. Shank, R.W. Schoenlein, D.M. Mittleman, J.J. Shiang,
and A.P. Alivisatos (With 4 Figures) .............................. 438

Quantum Beats Spectroscopy of Exciton Spin Dynamics
in GaAs Heterostructures
By S. Bar-Ad and I. Bar-Joseph (With 3 Figures) ..................... 443

Evidence of Slow Hole Spin Relaxation
in n-Modulation Doped GaAs/AlGaAs Quantum Well Structures
By Ph. Roussignol, P. Rolland, R. Ferreira, C. Delalande, G. Bastard,
A. Vinattieri, J. Martinez-Pastor, L. Carraresi, M. Colocci, J.F. Palmier,
and B. Etienne (With 1 Figure) .................................. 446

Femtosecond Time-Resolved Four-Wave Mixing in GaAs Quantum Wells
By D.S. Kim, J. Shah, T.C. Damen, J.E. Cunningham, W. Schäfer,
and S. Schmitt-Rink (With 4 Figures) .............................. 448

Exciton Radiative Lifetimes in GaAs Quantum Wells
By R. Eccleston, J. Kuhl, W.W. Rühle, and K. Ploog
(With 2 Figures) ............................................. 451
Optical Investigation of Bloch Oscillations in a Semiconductor Superlattice
(With 5 Figures) .............................................. 454

Coherent Pulse Breakup in Femtosecond Pulse Propagation in Semiconductors
By P.A. Harten, A. Knorr, S.G. Lee, R. Jin, F. Brown de Colstoun, E.M. Wright, G. Khitrova, H.M. Gibbs, S.W. Koch, and N. Peyghambarian (With 1 Figure) .................. 458

Absorption Saturation of the Urbach's Tail in Multiple Quantum Wells
By R. Raj, B.G. Sfez, D. Pellat, and J.L. Oudar (With 2 Figures) ............ 460

Photon Echo Polarisation Rules in GaAs Quantum Wells
By R. Eccleston, D. Bennhardt, J. Kuhl, P. Thomas, and K. Ploog (With 3 Figures) .............................................. 463

Observation of Many-Body Effects in the Femtosecond Temporal Profile of Quasi-2D Exciton Free-Induction Decay
By S. Weiss, M.-A. Mycek, J.-Y. Bigot, S. Schmitt-Rink, and D.S. Chemla (With 3 Figures) .................. 466

Radiative Recombination of Free Excitons in GaAs Quantum Wells
By B. Sermage, K. Satzke, C. Dumas, N. Roy, B. Deveaud, F. Clerot, and D.S. Katzer (With 4 Figures) ............ 472

Field-Enhanced GaAs/AlGaAs Waveguide Saturable Absorbers
By J.R. Karin, D.J. Derickson, R.J. Helkey, J.E. Bowers, and R.L. Thornton (With 2 Figures) .................. 475

Picosecond Excitonic Nonlinearities in the Presence of Disorder
By S.T. Cundiff and D.G. Steel (With 3 Figures) .................. 478

Fast Optical Nonlinearities in Semiconductor Quantum Dots
By G. Tamulaitis, R. Baltramiejūnas, S. Pakalnis, and A.I. Ekimov (With 2 Figures) .................. 482

Terahertz Radiation from Coherent Electron Oscillations in a Double-Quantum-Well Structure

Optical Generation of Terahertz Pulses from Polarized Excitons in Quantum Wells
By P.C.M. Planken and M.C. Nuss (With 3 Figures) .................. 487

Generation of High-Power Single-Cycle Picosecond Radiation
By D.R. Dykaar, R.R. Jones, D. You, D. Schumacher, and P.H. Bucksbaum (With 3 Figures) .................. 490

xix
Energetics and Dynamics of Global Protein Motion
By R.J.D. Miller, J. Deak, S. Palese, M. Pereira, L. Richard, and L. Schilling (With 2 Figures) ................. 525

Investigation of the Reaction Coordinate for Ligand Rebinding in Photoexcited Heme Proteins Using Transient Raman Spectroscopy
By H. Zhu, R. Lingle, Jr., X. Xu, and J.B. Hopkins
(With 2 Figures) .................................................. 528

Resonance Raman Studies of Electronic and Vibrational Relaxation Dynamics in Heme Proteins
By P.M. Champion, J.T. Sage, and P. Li ........................................ 533

Molecular Processes in the Primary Reaction of Photosynthetic Reaction Centers

Femtosecond Spontaneous Emission Studies of Photosynthetic Bacterial Reaction Centers
By S.J. Rosenthal, M. Du, X. Xie, T.J. DiMagno, M.E. Schmidt, J.R. Norris, and G.R. Fleming (With 1 Figure) .... 539

Subpicosecond Emission Studies of Bacterial Reaction Centers
By P. Hamm and W. Zinth (With 1 Figure) .................................. 541

Picosecond Fluorescence Kinetics of Purple Bacterial Reaction Centers
By M.G. Müller, K. Griebenow, and A.R. Holzwarth (With 2 Figures) .................................................. 543

Primary Radical Pair Formation in Photosystem-Two Reaction Centers

Energy Transfer and Primary Charge Separation in Heliobacteria by Picosecond Transient Absorption Spectroscopy
By P.I. van Noort, T.J. Aartsma, and J. Amesz (With 3 Figures) .......................................................... 549

Excitation Energy Transfer in Mutants of Rb. sphaeroides: The Effects of Changes in the Core Antenna Size
By L.M.P. Beekman, R.W. Visschers, K.J. Visscher, B. Althuis, W. Barz, D. Oesterhelt, V. Sundström, and R. van Grondelle (With 3 Figures) ............................................. 552

Femtosecond Excitation Transfer in Allophycocyanin
By A.V. Sharkov, E.V. Khoroshilov, I.V. Kryukov, P.G. Kryukov, T. Gillbro, R. Fischer, and H. Scheer (With 1 Figure) .................. 555

Femtosecond Förster Energy Transfer over 20 Å in Phycoerythrocyanin (PEC) Trimers
By L.O. Palsson, T. Gillbro, A. Sharkov, R. Fischer, and H. Scheer (With 1 Figure) .................................. 557

xxi
Ultrafast Energy Transfer Within the Light-Harvesting Antenna of Photosynthetic Purple Bacteria
By K.J. Visscher, V. Gulbinas, R.J. Cogdell, R. van Grondelle, and V. Sundström (With 2 Figures) ................................. 559

Femtosecond Dynamics in Rhodopsin
By T. Kobayashi, M. Taiji, K. Bryl, M. Nakagawa, and M. Tsuda (With 2 Figures) ................................................... 562

Subpicosecond Time-Resolved Spectroscopy of Halorhodopsin and Comparison with Bacteriorhodopsin
By H. Kandori, K. Yoshihara, H. Tomioka, H. Sasabe, and Y. Shichida (With 3 Figures) ............................................. 566

Part IX Chemistry: Electron and Energy Transfer, and Solvation Dynamics

Femtosecond Intermolecular Electron Transfer: Dye in Weakly Polar Electron-Donating Solvent
By K. Yoshihara, A. Yartsev, Y. Nagasawa, H. Kandori, A. Douhal, and K. Kemnitz (With 3 Figures) ......................... 571

Ultrafast Studies and Simulations on Direct Photoinduced Electron Transfer in the Betaines
By A.E. Johnson, N.E. Levinger, G.C. Walker, and P.F. Barbara (With 3 Figures) .................................................. 576

Picosecond Infrared Study of Ultrafast Electron Transfer and Vibrational Energy Relaxation in \([\text{NC}]_5\text{Ru}^{II}\text{CNRu}^{II}(\text{NH}_3)_5]^1-\)
By P.O. Stoutland, S.K. Doorn, R.B. Dyer, and W.H. Woodruff (With 1 Figure) ...................................................... 579

Ultrafast Studies on Intervalance Charge Transfer
By K. Tominaga, D.A.V. Kliner, J.T. Hupp, and P.F. Barbara (With 1 Figure) ...................................................... 582

Picosecond Infrared Study of Intramolecular Energy Transfer in \([(\text{phen})(\text{CO})_3\text{Re}^I(\text{NC})\text{Ru}^{II}(\text{CN})(\text{bpy})_2])^+\)
By R.B. Dyer, K.A. Peterson, K.C. Gordon, W.H. Woodruff, J.R. Schoonover, T.J. Meyer, and C.A. Bignozzi (With 1 Figure) .... 585

Noise-Induced Intramolecular Electron Transfer Processes in Polar Media
By P.O.J. Scherer ................................................................. 587

Femtosecond Proton Transfer in the Electronic Ground State of Vibrationally Hot Molecules
By T. Elsaesser, W. Frey, and M.T. Portella (With 2 Figures) .......... 589
Solvent Effects on the Fast Proton Transfer of 3-Hydroxyflavone
By B.J. Schwarz, L.A. Peteanu, and C.B. Harris (With 3 Figures) 592

Time-Resolved Charge Separation
in Acceptor-Substituted Anthrylpolyenes

Vibrationally Unrelaxed cis-Stilbene Photoproducts Examined Through Two-Color UV Pump-Probe Anti-Stokes Raman Spectroscopy
By D.L. Phillips, J.-M. Rodier, and A.B. Myers (With 4 Figures) 598

Vibrational Energy Redistribution and Relaxation
in the Photoisomerization of cis-Stilbene
By R.J. Sension, S.T. Repinec, A.Z. Szarka, and R.M. Hochstrasser (With 2 Figures) 601

Photoisomerization of cis-Stilbene in Compressed Solvents
By L. Nikowa, D. Schwarzer, J. Troe, and J. Schroeder (With 2 Figures) 603

Ultrafast Torsional Dynamics in Adsorbates: An SSHG Study
By M.J.E. Morgenthaler and S.R. Meech (With 1 Figure) 606

Barrierless Photochemical Isomerization
By U. Åberg, E. Åkesson, I. Fedchenia, and V. Sundström (With 2 Figures) 608

Femtosecond Molecular Dynamics in Liquids
By D.A. Wiersma, E.T.J. Nibbering, and K. Duppen (With 4 Figures) 611

Femtosecond Solvent Dynamics
Study by Time-Resolved Fluorescence and Transient Birefringence

Adiabatic and Nonadiabatic Effects in Solvation Dynamics
By E. Neria and A. Nitzan (With 1 Figure) 618

Excited-State Processes of 7-Azaindole
By M. Negrerie, F. Gai, J.-C. Lambry, J.-L. Martin, and J.W. Petrich (With 1 Figure) 621

Excited-State Proton Transfer and Hydrogen-Bonding Dynamics in 7-Azaindole: Time-Resolved Fluorescence and Computer Simulation
By C.F. Chapman, T.J. Marrone, R.S. Moog, and M. Maroncelli 624

Transient Hole Burning Studies of Electronic State Solvation:
Phonon and Structural Contributions
By J. Yu, J.T. Fourkas, and M. Berg (With 2 Figures) 626

xxiii
Subpicosecond Study of the Dynamic Processes in Push-Pull Styrenes and the Role of Solvation
By P. Hébert, G. Baldacchino, T. Gustavsson, V. Kabelka, P. Baldeck, and J.-C. Mialocq (With 3 Figures) 628

Picosecond Studies of Charge Transfer States in “Push-Pull” Linear Diphenyl Polyenes: Experimental Evidence for TICT and Bicimer States
By J.M. Viallet, F. Dupuy, R. Lapouyade, W.Q. Zheng, and C. Rullière (With 2 Figures) 631

Features of the Dual Fluorescence of 4-N,N-dialkylaminoalkylbenzoates in Alkanes
By M.C.C. de Lange, D.T. Leeson, A.H. Huizer, and C.A.G.O. Varma (With 1 Figure) 634

Investigation of Fast Relaxation Processes in Non-Fluorescent Rhodamine Dyes
By P. Plaza, N.D. Hung, M.M. Martin, Y.H. Meyer, and W. Rettig (With 1 Figure) 636

Femtosecond Photodissociation of Aromatic Disulfides Followed by Solvent Relaxation
By N.P. Ernsting (With 4 Figures) 638

Femtosecond Dynamics of C–O Bond Cleavage of a Spirooxazine Photochromic Reaction
By N. Tamai and H. Masuhara (With 2 Figures) 641

Dynamics of Molecular Rotation at the Air/Water Interface by Time-Resolved Second-Harmonic Generation
By A. Castro, D. Zhang, and K.B. Eisenthal (With 5 Figures) 644

Energy Relaxation and Redistribution in Large Molecules in Solution on Ultrafast Time Scales
By C.B. Harris, J.C. King, K.E. Schultz, B.J. Schwartz, and J.Z. Zhang (With 2 Figures) 650

Photodissociation and Recombination Dynamics of I₂ in Solution
By J.C. Alfano, D.A.V. Kliner, A.E. Johnson, N.E. Levinger, and P.F. Barbara (With 3 Figures) 653

Probing the Microscopic Molecular Environment in Liquids with Femtosecond Fourier-Transform Raman Spectroscopy
By D. McMorrow, S.K. Kim, J.S. Melinger, and W.T. Lotshaw (With 3 Figures) 656

The Homogeneity of Liquid Phase Vibrational Line Broadening from Raman Echo Experiments
By L.J. Muller, D. Vanden Bout, and M. Berg (With 2 Figures) 658
Excited State Photoreactions of Chlorine Dioxide in Solution
By R.C. Dunn and J.D. Simon (With 2 Figures) .................................. 661

Bimolecular Reactions are Power-Full
By A. Masad, S.Y. Goldberg, D. Huppert, and N. Agmon
(With 4 Figures) ................................................................. 664

Dynamics and Mechanism of Cu-Porphyrin Triplet Quenching
Through Liganding by Oxygen-Containing Solvents
By V.S. Chirvony and R. Gadonas ............................................. 667

Fast Processes in Liquid Alkane Photolysis
Above the Ionization Threshold
By M. Sander, U. Brummund, K. Luther, and J. Troe (With 1 Figure) .. 669

Index of Contributors ............................................................. 671
Femtosecond Excitation Transfer in Allophycocyanin

A.V. Sharkov$^1$, E.V. Khoroshilov$^1$, I.V. Kryukov$^1$, P.G. Kryukov$^1$, T. Gillbro$^2$, R. Fischer$^3$, and H. Scheer$^3$

$^1$P.N. Lebedev Physics Institute, Russian Academy of Sciences, 117924 Moscow, Russia  
$^2$Department of Physical Chemistry, University of Umea, S-901 87 Umea, Sweden  
$^3$Botanisches Institut der Universität München, W-8000 München, Fed. Rep. of Germany

Allophycocyanin (APC), C-phycocyanin (C-PC) and other biliproteins (phycoerythrin and phycoerythrocyanin) of cyanobacteria and red algae harvest solar energy in regions of the visible spectrum having weak chlorophyll absorption. This excitation energy is then efficiently transferred to chlorophyll in the photosynthetic membrane [1]. Light-harvesting complexes of cyanobacteria (phycobilisomes) consist of several hundred chromophores. The smallest (monomeric) subunit of C-PC consists of three (α-84, β-84 and β-155) chromophores. The APC monomeric subunit consists of two (α-84 and β-84) chromophores. In phycobilisomes APC and C-PC monomeric subunits are organised as trimers. Absorption spectrum of APC monomeric subunit changes drastically upon trimeric formation. Absorption maximum shifts from 620 nm for monomeric preparations to 650 nm with a 620-nm shoulder for trimeric preparations.

Isotropic absorption recovery kinetics with $\tau=440$ fs was recently observed in APC trimers at 615 nm [2-3]. The corresponding anisotropy was constant and close to 0.4 during the first few picoseconds. The results are consistent with a model of APC trimer in which α-84 and β-84 chromophores have different absorption spectra and femtosecond process corresponds to Förster energy transfer from 620-nm chromophore to neighbouring 650-nm one. Now new results from measurements in the 650-nm absorption band is presented here for further understanding of the ultrafast processes in APC trimers.

70-fs pulses at 620 nm from a CPM-laser were amplified at 100 Hz repetition rate in a multipass jet amplifier (pumping at 308 nm) and served as exciting pulses. A part of the amplified light was used for continuum generation. Pump-probe measurements were made with probe pulse polarization parallel or perpendicular to the polarization of the exciting pulse. APC was isolated from Mastigisladus laminosus.

Figure 1 shows difference spectra obtained in the 635-690 nm spectral region at parallel polarization of probe relative to the pump pulse. One can see that the bleaching of the 650-nm band is delayed relative to the bleaching of the 620-nm band. The strong red-shifted bleaching at 660 nm, due to stimulated emission, is further delayed relative to the bleaching of the 650-nm band. Fast recovery kinetics (corresponding to the 440-fs process observed at 615 nm [2-3]) was measured at 635 nm after initial bleaching (not shown). In contrast, only rise term was observed at 658 nm within the first picosecond after excitation. Decay of anisotropy from 0.4 to 0.2, however, occurs at that wavelength during this period. Femtosecond processes were not observed in monomeric preparations.

There are two possible explanations for the absorption spectrum of APC trimer. Due to the first one, 650-nm band and 620-nm shoulder both belong to α-84 and β-84 chromophores. One may believe that excitonic coupling between neighbouring α and β chromophores of different monomeric subunits is very strong (620-nm and 650-nm excitonic bands). For C-PC trimers the excitonic coupling (112 cm$^{-1}$) was calculated on the base of X-ray crystallography data [4]. Förster energy transfer between neighbouring excitonically coupled α and β chromophores with nearly similar absorption spectra was recently observed in C-PC trimers [5].
Unfortunately X-ray data are not available for APC trimers, but the excitonic splitting similar to C-PC could be assumed [6]. The 620-nm band in this case is a vibronic band of 650-nm electronic transition of α and β chromophores. An alternative explanation [3] is that one chromophore in a pair absorbs at 620 nm and the other (most likely β-84) at 650 nm due to a conformational change.

Our results strongly support the latter model. If the 650-nm band and the 620-nm shoulder belong to both chromophores, one should observe simultaneous bleaching of both bands due to ground state depletion. The delay in the bleaching of the 650-nm band ( 0 fs spectrum in comparison to the -150 fs spectrum ) corresponds to Förster energy transfer from a 620-nm donor to the 650-nm acceptor. Absorption recovery observed at 615 nm [2-3] and at 635 nm is due to relaxation of donor molecules to the ground state as a result of excitation energy transfer. Acceptor molecules absorbing mainly at 650 nm participate to a small extent to the isotropic or anisotropic kinetics measured at 615 nm [3]. The strong bleaching appearing near 660 nm is red-shifted relative to the bleaching of the 650-nm band ( 3 ps spectrum in comparison to 150 fs and 0 fs spectra). This shift is probably due to vibrational relaxation in the acceptor's excited state. Anisotropy decay observed at 658 nm is a result of Förster energy transfer between differently oriented donor and acceptor chromophores (as predicted from C-PC data).

References