Contents

<table>
<thead>
<tr>
<th>Part I</th>
<th>Overview and General Prospects</th>
</tr>
</thead>
</table>
|        | Additive Pulse Modelocking and Kerr-Lens Modelocking  
By H.A. Haus (With 6 Figures) | 3 |
|        | Molecular Control Spectrometer  
By Y. Yan, B.E. Kohler, R.E. Gillilan, R.M. Whitnell, K.R. Wilson, and S. Mukamel | 8 |
|        | Internal Motions of Proteins  
By M. Karplus | 13 |
|        | Some Theoretical Aspects of Electron Transfer in Supermolecules  
By J. Jortner and M. Bixon (With 3 Figures) | 15 |
|        | Femtosecond Time-Resolved Spectroscopy of Magneto-Excitons  
By D.S. Chemla, J.B. Stark, and W.H. Knox (With 6 Figures) | 21 |
|        | High-Order Harmonic Generation in Strong Laser Fields  
By A. L’Huillier and P. Balcou (With 3 Figures) | 29 |
|        | QED at $10^{20}$ W/cm$^2$  
By A.C. Melissinos (With 6 Figures) | 34 |

<table>
<thead>
<tr>
<th>Part II</th>
<th>Elementary Dynamics: Chemistry, Biology and Physics</th>
</tr>
</thead>
</table>
|        | Femtochemistry  
By A.H. Zewail (With 6 Figures) | 43 |
|        | Transient Dichroism Studies of I$_2$ Predissociation in Solution  
By N.F. Scherer, L.D. Ziegler, D. Jonas, and G.R. Fleming (With 3 Figures) | 49 |
|        | Investigation of the Primary Event in Vision  
Using 10 fs Blue-Green Optical Pulses  
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 Na$_2$
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 C$_{60}$

Femtosecond Dynamics of Molecular and Cluster Ionization and Fragmentation
By T. Baumert, R. Thalweiser, V. Weiß, 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
By L.D. Noordam, H. Stapelfeldt, D.I. Duncan, and T.F. Gallagher (With 3 Figures) ..................................................... 92
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
Single-Shot Measurement
of the Intensity and Phase of a Femtosecond Pulse
By D.J. Kane and R. Trebino (With 4 Figures) ................. 138

Two-Photon Interference Measurement of Ultrafast Laser Pulses
By M. Matsuoka, Y. Miyamoto, T. Kuga, M. Baba, and Y. Li
(With 2 Figures) ........................................... 140

Picosecond Single-Shot Pulse-Shape Measurement
by Stochastic Sampling of Detected Photon Times
By N. Adams, C. Bovet, E. Rossa, and A. Simonin (With 1 Figure) .... 142

Integrated Devices for Single Picosecond Pulse Measurements
By V. Gerbe, M. Cuzin, M.C. Gentet, and J. Lajzerowicz
(With 3 Figures) ........................................... 145

The C850X Ultrafast Streak Camera: An Instrument to Study Spatially
and Temporally Subpicosecond Laser–Matter Interaction
By A. Mens, R. Sauneuf, D. Schirmann, R. Verrecchia, P. Audebert,
J.C. Gauthier, J.P. Geindre, A. Antonetti, J.P. Chambaret, G. Hamoniaux,
and A. Mysyrowicz (With 2 Figures) .......................... 147

Distortion of a 6 fs Pulse in the Focus of a BK7 Lens
By Zs. Bor and Z.L. Horváth (With 1 Figure) .................... 150

Part IV Tools: Sources and Amplifiers

Modelocking, Stabilizing, and Starting Ultrashort Pulse Lasers
By E.P. Ippen (With 4 Figures) ................................ 155

17 fs Pulses from a Mode-Locked Ti:Sapphire Laser
By C.P. Huang, M.T. Asaki, S. Backus, H. Nathel, H.C. Kapteyn,
and M.M. Murnane (With 2 Figures) .......................... 160

Design Considerations for Femtosecond Ti:Sapphire Oscillators
By Ch. Spielmann, P.F. Curley, T. Brabec, E. Wintner, A.J. Schmidt,
and F. Krausz (With 3 Figures) ............................... 163

Self-Mode-Locked Cr³⁺:LiCaAlF₆ and Cr³⁺:LiSrAlF₆ Lasers
By A. Miller, P. Li Kam Wa, B.H.T. Chai, J.M. Evans, and W. Sibbett
(With 2 Figures) ........................................... 166

Sub-50 fs Pulse Generation
from a Self-Starting CW Passively Mode-Locked Cr:LiSrAlF₆ Laser
By N.H. Rizvi, P.M.W. French, and J.R. Taylor (With 2 Figures) .... 169

CW Krypton-Laser Pumped Cr³⁺:LiSrAlF₆ and Cr³⁺:LiSr₀.₈Ca₀.₂AlF₆
Crystals Produce 150 fs Mode-Locked Pulses
By A. Miller, P. Li Kam Wa, H.S. Wang, S.L. Ayres, E.W. Van Stryland,
and B.H.T. Chai (With 3 Figures) ............................. 172
60-fs Chromium-Doped Forsterite (Cr\textsuperscript{4+}:Mg\textsubscript{2}Si\textsubscript{4}O\textsubscript{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\textsubscript{2}O\textsubscript{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. Falliès, 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
Ultrafast Spectroscopy of Plasmas
Generated by Superintense Femtosecond Laser Pulses
By D. von der Linde, H. Schüler, H. Schulz, and T. Engers
(With 3 Figures) ................................. 280

Picosecond Soft-X-Ray Pulse Length Measurement
by Pump-Probe Absorption Spectroscopy
By M.H. Sher, U. Mohideen, H.W.K. Tom, O.R. Wood II,
G.D. Aumiller, D.L. Windt, W.K. Waskiewicz, J. Sugar, T.J. McClrath,
and R.R. Freeman (With 4 Figures) .......................... 283

Photon Acceleration via Laser-Produced Ionization Fronts
By R.L. Savage Jr., R.P. Brogle, W.B. Mori, and C. Joshi
(With 5 Figures) .................................. 286

Propagation of Intense Laser Pulses in Plasmas
By E. Esarey, P. Sprangle, J. Krall, and G. Joyce (With 1 Figure) .... 290

Ponderomotive Steepeening in Short-Scale-Length Laser-Plasmas
By D. Umstadter and X. Liu (With 2 Figures) .............................. 293

Possibility of Experimental Studies of Nonlinear Quantum
Electrodynamics Effects Using High Power Ultrashort Laser Pulses
By P.G. Kryukov (With 1 Figure) .................................. 296

Soliton-Like Self-Trapping of Three-Dimensional Patterns
By A. Barthelemy, C. Froehly, M. Shalaby, P. Donnat, J. Paye,
and A. Migus (With 9 Figures) ................................... 299

Physical Origins of the Spectral Continuum:
Self-Focusing, Self-Trapping and Cerenkov Radiation
By F. Salin, J. Watson, J.-F. Cormier, P. Georges, and A. Brun
(With 2 Figures) ...................................... 306

Diffraction and Focussing of Spectral Energy in a Two-Photon Process
By B. Broers, L.D. Noordam, and H.B. van Linden van den Heuvel
(With 3 Figures) ...................................... 309

Efficient Raman Conversion of Femtosecond UV Light Pulses
By K.A. Stankov and Y.-W. Lee (With 1 Figure) .......................... 311

Organic Crystalline Fiber
for Efficient Compression of Femtosecond Laser Pulses
By M. Yamashita (With 1 Figure) .................................. 313

Nonlinear Temporal Diffraction in Optical Fibers
By G.R. Boyer, M.K. Jackson, J. Paye, M.A. Franco, and A. Mysyrowicz
(With 3 Figures) ...................................... 315

Generation of a Soliton Pulse Train in an Optical Fibre
Using Two CW Single-Frequency Diode Lasers
By S.V. Chernikov, J.R. Taylor, P.V. Mamysev, and E.M. Dianov
(With 2 Figures) ...................................... 318
Experimental Investigation of Dark Solitons Interaction
By Ph. Emplit, J.-P. Hamaide, and M. Haelterman (With 3 Figures) .... 320

Femtosecond Pulse Propagation in Erbium-Doped Single-Mode Fibers
By J.M. Hickmann, A.S.L. Gomes, C.B. de Araújo, and A.S. Gouveia-Neto (With 3 Figures) ............... 323

Compression of Pulses from Soliton Fibre Lasers in a Dispersion-Decreasing Fibre
By S.V. Chernikov, D.J. Richardson, E.M. Dianov, and D.N. Payne
(With 4 Figures) ....................................................... 325

Part VI  Metals, Surfaces and Materials

Observation of the Thermalization of Electrons in a Metal Excited by Femtosecond Optical Pulses
By W.S. Fann, R. Storz, H.W.K. Tom, and J. Bokor
(With 2 Figures) ....................................................... 331

Femtosecond Thermionic Emission: Experiment, Analytical Theory, and Particle Simulations

Electron–Electron Dynamics Observed in Femtosecond Thermoreflection Measurements on Noble Metals
By R.H.M. Groeneveld, R. Sprik, and Ad. Lagendijk (With 2 Figures) .... 338

Inversion of Single- and Two-Photon Photocative Sensitivities of Metals in the Femtosecond Range
By J.P. Girardeau-Montaut, C. Girardeau-Montaut, S.D. Moustazi, and C. Fotakis (With 1 Figure) ............... 340

Femtosecond Relaxation of Plasma Excitations in Silver Films
By R.A. Höpfel, D. Steinmüller-Nethl, F.R. Aussenegg, and A. Leitner
(With 3 Figures) ....................................................... 342

Femtosecond Free Induction Decay of Metal Surface Adsorbate Vibrations
By 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) .... 345

Observation of Laser-Induced Desorption of CO from Cu(111) with 100 fs Time-Resolution
By J.A. Prybyla, H.W.K. Tom, and G.D. Aumiller (With 2 Figures) .... 347

Femtosecond Desorption of Molecular Oxygen from Pt(111)
By F.-J. Kao, D.G. Busch, D. Gomes da Costa, D. Cohen, and W. Ho
(With 1 Figure) ....................................................... 350
Femtosecond Carrier Dynamics in Solid C₆₀ 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-SiO₂
By W. Joosen, S. Guizard, P. Martin, G. Petite, P. Agostini,
A. Dos Santos, G. Grillon, J.P. Chambaret, D. Hulin, A. Migus,
and A. Antonetti (With 4 Figures) ................................ 362

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 PbTiO₃:
A₁ 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
By A. Cybo-Ottone, M. Nisoli, V. Magni, S. De Silvestri, O. Svelto,
G. Zerbi, and R. Tubino (With 2 Figures) .......................... 370

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

xvi
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 C60 and C70 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

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. Ecclestone, 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
Transient Electron Transport in GaAs Quantum Wells: From the Ballistic to the Quasi-Equilibrium Regime
By W. Sha, J. Rhee, and T.B. Norris (With 4 Figures) .......... 493

A Novel Free-Standing Absolute-Voltage Probe with 2.3-Picosecond Resolution and 1-Microvolt Sensitivity
By J. Kim, S. Williamson, J. Nees, and S. Wakana
(With 3 Figures) ........................................... 496

Picosecond Pseudomorphic AlGaAs/InGaAs MODFET Large-Signal Switching Measured by Electro-Optic Sampling
(With 3 Figures) ........................................... 500

Ultrafast Decay of Photodiffractive Gratings in Hetero n-i-p-i’s by Enhanced In-Plane Transport

Picosecond High-Sensitivity In_xGa_1-xAs Photodetectors
By S. Gupta, J.F. Whitaker, S.L. Williamson, P. Ho, J.S. Mazurowski, and J.M. Ballingall (With 2 Figures) .................. 505

An Ultrafast Polarization-Independent All-Optical Demultiplexer Utilizing Induced-Frequency Shift
By T. Morioka, K. Mori, and M. Saruwatari (With 2 Figures) .... 508

Electrical Soliton Devices as >100 GHz Signal Sources
By E. Carman, M. Case, M. Kamegawa, R. Yu, K. Giboney, and M. Rodwell (With 2 Figures) ...................... 511

Determination of Photonic Band Gaps and Dispersion in Two-Dimensional Dielectric Arrays with Ultrafast Electromagnetic Transients
By W.M. Robertson, G. Arjavalingam, R.D. Meade, K.D. Brommer, A.M. Rappe, and J.D. Joannopoulos (With 2 Figures) ........ 513

Part VIII  Biology: Primary Dynamics, Electron and Energy Transfer

Ultrafast Infrared Spectroscopy of Protein Dynamics

Ultrafast Near-IR Spectroscopy of Carbonmonoxymyoglobin: The Dynamics of Protein Relaxation
By M. Lim, T.A. Jackson, and P.A. Anfinrud (With 4 Figures) .... 522

xx
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
By W. Zinth, C. Lauterwasser, U. Finkele, P. Hamm, S. Schmidt, and W. Kaiser (With 3 Figures) .......................... 535

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
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 [(NC)₅Ru⁺CNRu⁺(NH₃)₅]⁻
By P.O. Stoutland, S.K. Doorn, R.B. Dyer, and W.H. Woodruff (With 1 Figure) ................................................................. 579

Ultrafast Studies on Intervalence 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 [(phen)(CO)₃Re⁺(NC)Ru⁺(CN)(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
By H. Port, G. Quapil, H.C. Wolf, F. Effenberger, C.-P. Niesert,
R. Buhleier, Z. Gogolak, and J. Kuhl (With 2 Figures) ...................... 596

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
Studied by Time-Resolved Fluorescence and Transient Birefringence
By S.J. Rosenthal, N.F. Scherer, M. Cho, X. Xie, M.E. Schmidt,
and G.R. Fleming (With 2 Figures) ...................................... 616

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. Négrerie, 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
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. Miallocq (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

XXIV
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 Förster Energy Transfer over 20 Å in Phycoerythrocyanin (PEC) Trimers

L.O. Palsson¹, T. Gillbro¹, A. Sharkov¹, R. Fischer², and H. Scheer²

¹Department of Physical Chemistry, University of Umeå, S-901 87 Umeå, Sweden
²Botanisches Institut der Universität München, W-8000 München, Fed. Rep. of Germany

Energy transfer between violobilin (α-84) and phycocyanin (β-84) chromophores in phycoerythrocyanin trimers from Mastigocladus laminosus occurs in 400 fs indicating a Förster-type energy transfer over 20 Å.

Introduction.

Phycobilisomes constitute the antenna for capture of light energy and are active in the subsequent transfer of the excitation energy to the reaction centers in red algae and cyanobacteria. The phycobilisome contains chromophores of several different types and thus have the capacity to absorb light of different wavelengths. This enables the organism to harvest light energy and grow in different radiation environments. Thus far only a few photosynthetic antenna complexes have been crystallized and their structure determined. One example of a highly resolved structure is the phycoreythrocyanin antenna complex isolated from phycobilisomes of Mastigocladus laminosus [1]. In this work the positions and orientations of the three chromophores (α-84, β-84 and β-155) were determined. The closest contact is between the α-84 (violobilin) and β-84 (phycocyanin) chromophores with a center-to-center distance of 19 Å. The α-chromophores is supposed to absorb strongly about 572 nm, while the β-subunit absorbs at longer wavelengths, i.e. about 592 nm [2].

Results and discussion.

Exciting at 575 nm using cavity dumped compressed pulses, FWHM ~ 50 fs we resolve a very fast 400 fs partial recovery of the bleaching signal. This indicates an ultrafast energy transfer from α-84 to a red-shifted β-chromophore. The process we observe is actually an equilibration of the excitation energy between these chromophores. A
Fig. Absorption recovery at 575 nm and at 600 nm.

similar but reversed 400 fs process is observed if we excite at 600 nm (mainly β-chromophores). In this case a slow recovery of the transient bleaching is estimated to occur in about 50 ps is observed as well. From the crystal structure and an estimated Förster radius ($R_o$) of 60-70 Å for this sub picosecond energy transfer we calculate a transfer rate, $k$, of

$$k = \frac{K^2}{\tau} \left( \frac{R_o}{R} \right)^6 = 978 \text{ ns}^{-1}$$

where $\tau = 2.0$ ns, $K = -1.10$, $R_o = 65$ Å and $R = 19$ Å. For the equilibration process $\Sigma k = 1950$ ns$^{-1}$, giving a lifetime of about 500 fs in fair agreement with the experimental results. The results reported here are also consistent with recent work on allophycocyanin and c-phycocyanin trimers [3,4].

References:

1. M. Duerring, R Huber, W. Bode, R. Ruembeli and H. Zuber
2. R. Füglistaller, F. Suter and H.Zuber