Ultrafast Phenomena V

Proceedings of the Fifth OSA Topical Meeting
Snowmass, Colorado, June 16–19, 1986

Editors: G. R. Fleming and A. E. Siegman

With 427 Figures

Springer-Verlag Berlin Heidelberg New York
London Paris Tokyo
Contents

Part I Mode Locking and Ultrashort Pulse Generation

Passive and Hybrid Femtosecond Operation of a Linear Astigmatism Compensated Dye Laser
By J.-C. Diels, N. Jamasbi, and L. Sarger (With 1 Figure) ........... 2

By M.D. Dawson, T.F. Boggess, D.W. Garvey, and A.L. Smirl (With 4 Figures) ........................................ 5

Cavity-Mirror Dispersion Dependence of Pulse Duration Generated from a Simple CPM Laser: An Experimental Study
By M. Yamashita, K. Torizuka, T. Sato, and M. Ishikawa (With 2 Figures) ............................................. 8

Femtosecond Pulse Generation from Passively Mode Locked Continuous Wave Dye Lasers 550–700 nm
By P.M.W. French and J.R. Taylor (With 3 Figures) ............... 11

Stabilisation of a CPM Dye Laser Synchronously Pumped by a Frequency Doubled ML YAG Laser
By J. Chesnoy and L. Fini (With 3 Figures) ....................... 14


Experimental Observation of High Order Solitons in a Colliding Pulse Mode-Locked Laser
By F. Salin, P. Grangier, G. Roger, and A. Brun (With 4 Figures) 20

Advances in the Theory of Mode-Locking by Synchronous Pumping. By G.H.C. New and J.M. Catherall (With 3 Figures) ... 24

Collective Modes - An Analytical Model for Active Mode Locking in the Transient Case
By P. Aechtner, P. Heinz, and A. Laubereau (With 2 Figures) .... 27
Generation of Picosecond Pulses from a Continuous Wave Neodymium:Phosphate Glass Laser
By L. Yan, J.D. Ling, P.-T. Ho, and C.H. Lee (With 3 Figures) .... 30

Part II Ultrafast Optical Generation and Measurement Techniques


New Optical Design for a Jet Amplifier. By O. Seddiki, A. Goddi, R. Mounet, J.-F. Morhange, and C. Hirlimann (With 2 Figures) ... 43

Fiber Raman Amplification Soliton Laser (FRASL)
By M.N. Islam, L.F. Mollenauer, and R.H. Stolen (With 4 Figures) 46

Dispersion Compensated Fiber Raman Oscillator
By J.D. Kafka, D.F. Head, and T. Baer (With 2 Figures) .......... 51

80-fs Soliton-like Pulses from an Optical Nonlinear Fiber Resonator. By B. Zysset, P. Beaud, W. Hödel, and H.P. Weber (With 4 Figures) ................................................. 54

The Stabilized Soliton Laser
By F.M. Mitschke and L.F. Mollenauer (With 4 Figures) .......... 58

The Soliton Self Frequency Shift. By F.M. Mitschke, L.F. Mollenauer, and J.P. Gordon (With 2 Figures) ............. 62

Solitons at the Zero Dispersion Wavelength of Single-Mode Fibers
By P.K.A. Wai, C.R. Menyuk, H.H. Chen, and Y.C. Lee (With 1 Figure) ................................................................. 65

Active Mode-Locking of an InGaAsP Optical-Fiber Ring Laser
By G. Eisenstein, R.M. Jopson, M.S. Whalen, K.L. Hall, and G. Raybon (With 4 Figures) ....................... 68

Femtosecond Resolved Fluorescence
By W. Rudolph and J.-C. Diels (With 2 Figures) .............. 71

Measurement of Optical Phase with Subpicosecond Resolution by Time Domain Interferometry. By J.E. Rothenberg (With 6 Figures) .......................................................... 78

Real Time Picosecond Optical Oscilloscope
By J.A. Valdmanis (With 8 Figures) ........................................... 82

Beam Overlap for Long Delay Lines Using Active Feedback
By C. Doland, W.B. Jackson, and A. Andersson (With 3 Figures) ... 86


An Investigation on Ultrashort Light Pulse Generation by Travelling-Wave Amplified Spontaneous Emission
By W. Lee, C. Ning, Z. Huang, and W. Wang (With 5 Figures) ... 92

Part III Electrooptic Sampling Techniques

Electrooptic Sampling of Gallium Arsenide Integrated Circuits


Precise Measurement of Signal Propagation Characteristics in GaAs Integrated Circuits by Picosecond Electro-Optic Sampling
By R.K. Jain, X.-C. Zhang, M.G. Ressl, and T.J. Pier (With 2 Figures) .......................................................... 107

Propagation of Ultrashort Electrical Pulses on Superconducting Transmission Lines
By I.N. Duling III, C.-C. Chi, W.J. Gallagher, D. Grischkowsky, N.J. Halas, M.B. Ketchen, and A.W. Kleinsasser (With 4 Figures) 110


Picosecond Optoelectronic Sampling of Electrical Waveforms Produced by an Optically Excited Field Effect Transistor
By D.E. Cooper and S.C. Moss (With 1 Figure) ......................... 117
<table>
<thead>
<tr>
<th>Title</th>
<th>Authors</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>High Speed Circuit Measurements Using Photoemission Sampling</td>
<td>J. Bokor, A.M. Johnson, R.H. Storz, and W.M. Simpson</td>
<td>123</td>
</tr>
<tr>
<td>Nonlinear Responses of Picosecond Photodetectors to Photogenerated Carriers</td>
<td>T.F. Caruthers and J.F. Weller</td>
<td>131</td>
</tr>
<tr>
<td>Elimination of Dynamic Flash in a Picosecond Streak Image Tube</td>
<td>Huanwen Zhang</td>
<td>137</td>
</tr>
<tr>
<td>Part IV Nonlinear Optics and Continuum Generation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>New Excitation and Probe Continuum Sources for Subpicosecond Absorption Spectroscopy</td>
<td>J.H. Glownia, J. Misewich, and P.P. Sorokin</td>
<td>153</td>
</tr>
<tr>
<td>Induced Phase Modulation and Spectral Broadening of a Weak 530-nm Picosecond Pulse by an Intense 1060-nm Picosecond Pulse in Glass.</td>
<td>R.R. Alfano, Q.X. Li, T. Jimbo, J.T. Manassah, and P.P. Ho</td>
<td>157</td>
</tr>
<tr>
<td>The Observation of Chirped Stimulated Raman Scattered Light in Fibers.</td>
<td>A.M. Johnson, R.H. Stolen, and W.M. Simpson</td>
<td>160</td>
</tr>
</tbody>
</table>
Observation of 7.2-THz Beats Between the D-Lines of Atomic Rb
By J.E. Golub and T.W. Mossberg (With 1 Figure) .......................... 164

Coherent Multiphoton Resonant Interaction and Harmonic Generation. By A. Mukherjee, N. Mukherjee, J.-C. Diels, and G. Arzumanyan (With 4 Figures) .......................... 166

Ultrafast Chaos from Semiconductor Lasers. By Y. Cho, T. Umeda, I. Jun Cha, M. Koishi, and M. Miwa (With 5 Figures) . 169

Thermodynamics and Kinetics of Melting, Evaporation and Crystallization, Induced by Picosecond Pulsed Laser Irradiation
By F. Spaepen (With 1 Figure) ......................................................... 174


Superheating During Ultrafast Laser Heating of Semiconductors
By D. von der Linde, N. Fabricius, B. Danielzik, and P. Hermes (With 4 Figures) ................................................................. 182

Non-equilibrium Carriers in GaAs: Secondary Emission During the First Two Picoseconds
By J.A. Kash and J.C. Tsang (With 5 Figures) ......................... 188

Ultrafast Carrier Dynamics in GaAs and AlxGa1-xAs
By W.Z. Lin, J.G. Fujimoto, E.P. Ippen, and R.A. Logan (With 4 Figures) ................................................................. 193


Picosecond Relaxation of Nonthermal Wannier Excitons in GaAs
By L. Schultheis, J. Kuhl, A. Honold, and C.W. Tu (With 1 Figure) ................................................................. 201

Picosecond Observation of the Photorefractive Effect in GaAs
By A.L. Smirl, G.C. Valley, M.B. Klein, K. Bohnert, and T.F. Boggess (With 2 Figures) ................................................................. 203
Time-Resolved Photoluminescence Measurements in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ Under Intense Picosecond Excitation

Picosecond Excite-Probe and Transient Grating Studies of $\text{Ga}_x\text{In}_{1-x}\text{As}_y\text{P}_{1-y}$. By R.J. Manning, A. Miller, A.M. Fox, and J.H. Marsh (With 2 Figures) ........................................... 210

Ultrafast Dynamics in GaAlAs Diode Laser Amplifiers
By M.S. Stix, M.P. Kesler, and E.P. Ippen (With 8 Figures) .......... 213

Electronic Energy Relaxation and Localization in Two II-VI Compound Semiconductor Quantum Well Structures
By Y. Hefetz, W.C. Goltsos, D. Lee, and A.V. Nurmikko (With 6 Figures) ................................................................. 218

Transient Raman Scattering in Multiple Quantum Well Structures
By D.Y. Oberli, D.R. Wake, M.V. Klein, J. Klem, and H. Morkoç (With 2 Figures) ........................................................... 223

Fast Energy Relaxation of Hot Electrons in Bulk GaAs and Multi-Quantum Wells. By C.H. Yang and S.A. Lyon (With 2 Figures) ... 227

Picosecond Photoluminescence and Energy-Loss Rates in GaAs Quantum Wells Under High-Density Excitation
By T. Kobayashi, H. Uchiki, Y. Arakawa, and H. Sakaki (With 4 Figures) ................................................................. 231

Broad Tuning of the Photoluminescence Energy and Lifetime by the Quantum-Confined Stark Effect. By H.-J. Polland, L. Schultheis, J. Kuhl, E.O. Göbel, and C.W. Tu (With 2 Figures) 234

Auger Heating of Silicon-on-Sapphire by Femtosecond Optical Pulses. By M.C. Downer and C.V. Shank (With 2 Figures) ........ 238

The Origin of Picosecond Photoinduced Absorption Decays in Hydrogenated Amorphous Silicon
By W.B. Jackson, C. Doland, and C.C. Tsai (With 2 Figures) ...... 242

Picosecond Decay of Photoinduced Absorption in Hydrogenated Amorphous Silicon
By D.M. Roberts and T.L. Gustafson (With 2 Figures) .................. 245

Femtosecond Spectroscopy of Hot Carriers in Germanium
By P.M. Fauchet, D. Hulin, G. Hamoniaux, A. Orszag, J. Kolodzey, and S. Wagner (With 3 Figures) ...................................... 248
Spin Dephasing Kinetics of Free Carriers in Alloy Semimagnetic Semiconductors \(\text{Cd}_{1-x}\text{Mn}_x\text{Se}\) by One and Two Photon Excitation
By M.R. Junnarkar and R.R. Alfano (With 1 Figure) ..................... 251

Detection of Higher Order Fourier Components of Index Gratings in Picosecond Transient Grating Experiments
By E.O. Göbel and H. Saito (With 3 Figures) ....................... 254

Transient Thermoreflectance Studies of Thermal Transport in Compositonally Modulated Metal Films. By G.L. Eesley, C.A. Paddock, and B.M. Clemens (With 2 Figures) ................. 257


Femtosecond Carrier Relaxation in Semiconductor-Doped Glasses
By M.C. Nuss, W. Zinth, and W. Kaiser (With 2 Figures) ........... 267


High-Contrast Ultrafast Phase Conjugation in Semiconductor-Doped Glass. By D. Cotter (With 3 Figures) ......................... 274

Femtosecond Vibrational Relaxation of the \(\text{F}^+\) Center in LiF
By W.H. Knox, L.F. Mollenauer, and R.L. Fork (With 2 Figures) . 277

Determination of the Rapid Quenching Rates of Excited State F-Centers by \(\text{OH}^-\) Defects in KCl. By Du-Jeon Jang, T.C. Corcoran, M.A. El-Sayed, L. Gomes, and F. Luty (With 4 Figures) ................................. 280

Propagation of Coherent Phonon Polaritons in \(\text{LiTaO}_3\) Measured by FIR-Cherenkov-Pulses
By M.C. Nuss and D.H. Auston (With 3 Figures) .................... 284

Part VI Chemical Reaction Dynamics

Cages, Crossings and Correlations – Theoretical Perspectives on Solution Reaction Dynamics. By J.T. Hynes ......................... 288
Polarity Dependent Barriers and the Photoisomerization Dynamics of Polar Molecules in Solution. By J.M. Hicks, M.T. Vandersall, E.V. Sitzmann, and K.B. Eisenthal (With 3 Figures) .................. 293

Dynamic Solvent Effects on Small Barrier Isomerizations
By P.F. Barbara and V. Nagarajan (With 3 Figures) ................. 299

Solvation Dynamics in Polar Liquids: Experiment and Simulation
(With 4 Figures) .................................................. 303


Time-Dependent Fluorescence Shift in Alcoholic Solvents:
A Non-Debye Behaviour Related to Hydrogen Bonds
By C. Rullière, A. Declémy, and Ph. Kottis (With 4 Figures) ....... 312

Picosecond Dynamics of Proton-Anion Ion Pair Geminate Recombination. By D. Huppert and E. Pines (With 1 Figure) ...... 315

Excited State Proton Transfer in Matrix Isolated Water and Methanol Complexes of 2-Hydroxy-4,5-benzotropone and 3-Hydroxyflavone
By D.F. Kelley and G.A. Brucker (With 2 Figures) .................. 319

Detection of the Inverted Region in Photo-induced Intramolecular Electron Transfer. By R.J. Harrison, G.S. Beddard, J.A. Cowan, and J.K.M. Sanders (With 2 Figures) ......................... 322

Ultrafast Studies Designed to Test the Fundamental Statistical Assumptions Underlying Chemical Reactivity in Liquids
By C.B. Harris, J.K. Brown, M.E. Paige, D.E. Smith, and D.J. Russell (With 4 Figures) ................................. 326

Geminate Recombination and Relaxation of Condensed Phase Molecular Halogens
By D.F. Kelley and N.A. Abul Haj (With 1 Figure) ................... 330

(With 4 Figures) .................................................. 334

Cage Recombination and Unimolecular β-Scission Reactions of Sulfur Centered Free Radicals
By T.W. Scott and S.N. Liu (With 3 Figures) ......................... 338

Kramers-Hubbard Approach to the Solvent Dependence of Isomerization. By M. Lee and R.M. Hochstrasser (With 3 Figures) 344

Photoisomerization Studies of Substituted Stilbenes: 4,4'-Dihydroxystilbene and 4,4'-Dimethoxystilbene By D.M. Zeglinski and D.H. Waldeck (With 2 Figures) ............... 347

Picosecond Studies of Barrierless Torsional Diffusion By D. Ben-Amotz and C.B. Harris (With 2 Figures) ..................... 350

Time-Resolved Fluorescence Spectra of Ethidium Bromide By J.H. Sommer, T.M. Nordlund, M. McGuire, and G. McLendon (With 3 Figures) .......................................................... 353

Picosecond and Femtosecond Molecular Beam Chemistry: Coherence and Fragment Recoil Dynamics By A.H. Zewail (With 4 Figures) ........................................ 356

Picosecond Laser Study of the Collisionless UV Photodissociation of Energetic Materials By J.-C. Mialocq and J.C. Stephenson (With 3 Figures) ................. 362


Time-Resolved Measurement of Laser-Induced Desorption of a Molecular Monolayer. By G. Arjavalingam, T.F. Heinz, and J.H. Glownia (With 1 Figure) .............................................. 370

Part VII Dynamics of Biological Processes


Femtosecond Spectroscopy of the Primary Events of Bacterial Photosynthesis By W. Zinth, J. Dobler, and W. Kaiser (With 3 Figures) .......... 379

An Accumulated Photon Echo Study of Sub-picosecond Processes in Photosynthetic Reaction Centers By S.R. Meech, A.J. Hoff, and D.A. Wiersma (With 2 Figures) .... 384
Ultrafast Electron and Energy Transfer in Reaction Center and Antenna Proteins from Photosynthetic Bacteria
By M.R. Wasielewski, D.M. Tiede, and H.A. Frank
(With 5 Figures) ......................................................... 388

Femtosecond Spectroscopy of Excitation Energy Transfer and Initial Charge Separation in the Reaction Center of the Photosynthetic Bacterium Rhodopseudomonas sphaeroides
By J. Breton, J.-L. Martin, A. Migus, A. Antonetti, and A. Orszag
(With 4 Figures) ......................................................... 393

Picosecond Transient Absorption Spectroscopy of Green Plant Photosystem I Reaction Centres
By B.L. Gore, L.B. Giorgi, and G. Porter (With 1 Figure) .......... 398

Femtosecond-Pulse Spectroscopy of Primary Photoprocesses in Reaction Centers of Rhodopseudomonas sphaeroides R-26
By S.V. Chekalin, Yu.A. Matveets, and A.P. Yartsev
(With 3 Figures) ......................................................... 402

(With 1 Figure) ......................................................... 406


(With 3 Figures) ......................................................... 413

Primary Process of Vision: Hypsorhodopsin
By T. Kobayashi, H. Ohtani, and M. Tsuda (With 3 Figures) ...... 416

Reactivity and Dynamics of Hemeproteins in the Femtosecond and Picosecond Time Domains. By D. Houde, J.W. Petrich, O.L Rojas, C. Poyart, A. Antonetti, and J.L. Martin (With 3 Figures) ...... 419

Picosecond Raman Hole Burning as a Probe of Conformational Heterogeneity: Applications to Oxyhemoglobin
By B.F. Campbell and J.M. Friedman (With 4 Figures) .......... 423


Molecular Dynamics Study of Vibrational Cooling in Optically Excited Hemeproteins. By E.R. Henry, W.A. Eaton, and R.M. Hochstrasser (With 1 Figure) .......... 430

XVI

Part VIII  Energy Transfer and Relaxation


Optical Pump-Probe Spectroscopy of Dyes on Surfaces: Ground-State Recovery of Rhodamine 640 on ZnO and Fused Silica By P.A. Anfinrud, T.P. Causgrove, and W.S. Struve (With 1 Figure) ......................................................... 442

Picosecond Fluorescence Spectroscopy on Molecular Association in Langmuir-Blodgett Films By I. Yamazaki, N. Tamai, and T. Yamazaki (With 3 Figures) ...... 444

Fluorescence Concentration Depolarization of DODCI in Glycerol: A Photon-Counting Test of Three-Dimensional Excitation Transport Theory By D.E. Hart, P.A. Anfinrud, and W.S. Struve (With 1 Figure) ... 447

Fractal Behaviors in Two-Dimensional Excitation Energy Transfer on Vesicle Surfaces. By N. Tamai, T. Yamazaki, I. Yamazaki, and N. Mataga (With 3 Figures) ................................................. 449


Nonlinear Absorption Spectroscopy of Liquids with Ultrashort IR Pulses By H. Graener, R. Dohlus, and A. Laubereau (With 2 Figures) .... 458

Femtosecond Relaxation Dynamics of Large Organic Molecules By M.J. Rosker, F.W. Wise, C.L. Tang, and A.J. Taylor (With 4 Figures) ............................................................. 461

Population Lifetimes of OH(v=1) and OD(v=1) Vibrations in Alcohols, Silanols and Crystalline Micas. By E.J. Heilweil, M.P. Casassa, R.R. Cavanagh, and J.C. Stephenson ......................... 465

$S_0$-$S_n$ Two-Photon Absorption Dynamics of Rhodamine Dyes By P. Sperber, M. Weidner, and A. Penzkofer (With 3 Figures) .... 469

Picosecond Photoconductivity and Nonlinear Optical Phenomena in trans-Polyacetylene
By D. Moses, M. Sinclair, and A.J. Heeger (With 1 Figure) ........ 475

Singlet Exciton Fusion in Molecular Solids
By R.R. Millard and B.I. Greene (With 2 Figures) ...................... 478

Matrix Effect on Vibrational Relaxation in Molecular Crystals
By J.R. Hill, E.L. Chronister, J.C. Postlewaite, and D.D. Dlott (With 1 Figure) ............................................. 482

Optical Damage in Molecular Crystals: A Solid State Explosion
By D.D. Dlott, T.J. Kosic, and J.R. Hill (With 4 Figures) .......... 485

Rotational Relaxation of Free and Solvated Rotors
By A.J. Bain, C. Han, P.L. Holt, P.J. McCarthy, A.B. Myers, M.A. Pereira, and R.M. Hochstrasser (With 5 Figures) .......... 489

Ultrafast Dynamics at the Interface: Probing the Transition from Solution to Surface Interactions in Charged Micelles
By E.F. Templeton, K. Brinker, S. Paone, and G.A. Kenney-Wallace (With 1 Figure) ............................................. 495

Shock Moderated Photophysics and Photochemistry at Multi-kilobar Pressures
By B.L. Justus, A.L. Huston, and A.J. Campillo (With 5 Figures) . 499

Part IX Coherent Spectroscopic Techniques

Phase Grating Approach to Susceptibility Tensors: Determination in Isotropic Media. By J. Etchepare, G. Grillon, I. Thomazeau, G. Hamoniaux, and A. Orszag (With 4 Figures) ....................... 504

Nonlinear Response Function for Four-Wave Mixing: Application to Coherent Raman Lineshapes in Polyatomics and to the Optical Anderson Transition
By S. Mukamel, Z. Deng, and R.F. Loring (With 2 Figures) ........ 510

Picosecond Laser Pulse Shaping and Phase Shifting for Molecular Spectroscopy
By M. Haner, F. Spano, and W.S. Warren (With 2 Figures) ........ 514


XVIII
<table>
<thead>
<tr>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Picosecond Raman-Induced Phase Conjugation Spectroscopy</td>
<td>521</td>
</tr>
<tr>
<td>By R. Dorsinville, P. Delfyett, and R.R. Alfano (With 2 Figures)</td>
<td></td>
</tr>
<tr>
<td>Polarization Dependence of Time-Resolved CARS in Liquids</td>
<td>524</td>
</tr>
<tr>
<td>By N. Kohles and A. Laubereau (With 3 Figures)</td>
<td></td>
</tr>
<tr>
<td>Direct Measurement of Wave-Vector-Dependent Polariton Energy</td>
<td>528</td>
</tr>
<tr>
<td>Velocity and Dephasing in NH₄Cl</td>
<td></td>
</tr>
<tr>
<td>By G.M. Gale, F. Vallée, and C. Flytzanis (With 4 Figures)</td>
<td></td>
</tr>
<tr>
<td>Impulsive Stimulated Rayleigh, Brillouin, and Raman Scattering:</td>
<td>532</td>
</tr>
<tr>
<td>Experiments and Theory of Light Scattering Spectroscopy in the</td>
<td></td>
</tr>
<tr>
<td>Time Domain. By M.R. Farrar, L.R. Williams, Yong-Xin Yan,</td>
<td></td>
</tr>
<tr>
<td>Lap-Tak Cheng, and K.A. Nelson (With 4 Figures)</td>
<td></td>
</tr>
<tr>
<td>Ultrafast Transient Spectroscopy with Broadband Non-Transform-</td>
<td>536</td>
</tr>
<tr>
<td>Limited Light Sources</td>
<td></td>
</tr>
<tr>
<td>By T. Yajima and N. Morita (With 4 Figures)</td>
<td></td>
</tr>
<tr>
<td>Picosecond Dephasing Time Measurement by CSRS Using</td>
<td>541</td>
</tr>
<tr>
<td>Temporally Incoherent Nanosecond Laser with Short Correlation</td>
<td></td>
</tr>
<tr>
<td>Time</td>
<td></td>
</tr>
<tr>
<td>By T. Kobayashi, T. Hattori, and A. Terasaki (With 3 Figures)</td>
<td></td>
</tr>
<tr>
<td>Anomalous Pulse Duration Dependence of the Quasicontinuum Absorption</td>
<td>544</td>
</tr>
<tr>
<td>Spectrum</td>
<td></td>
</tr>
<tr>
<td>By P. Mukherjee and H.S. Kwok (With 3 Figures)</td>
<td></td>
</tr>
<tr>
<td>Index of Contributors</td>
<td>549</td>
</tr>
</tbody>
</table>
Femtosecond Carrier Relaxation in Semiconductor-Doped Glasses

M.C. Nuss*, W. Zinth, and W. Kaiser

Physik Department der Technischen Universität München, Arcisstr. 21, D-8000 München 2, Fed. Rep. of Germany

Semiconductor-doped glasses incorporating small CdS\textsubscript{x}Se\textsubscript{1-x} or CdSe\textsubscript{x}Te\textsubscript{1-x} crystallites in a glass matrix, show strong nonlinear absorption /1/ and large values of the nonlinear optical susceptibility \( \chi^3 \) /2/. The semiconductor crystallite system currently attracts increasing attention when the semiconductor inclusions are small enough to expect quantum effects.

The semiconductor-doped glass filters studied (Schott RG 830, RG 715, and RG 645) are characterized by an exponential absorption edge ~ three times less steep than the corresponding bulk semiconductors. The frequency position of the absorption edge varies with composition, but it may as well be influenced by the size of the semiconductor inclusions. No confinement effects are observed in the absorption spectra at 300 K, most probably due to a certain variation of crystallite size.

Femtosecond absorption recovery of all three Schott filter glasses was studied. In the experiment, femtosecond light pulses of \( \approx 60 \) fs duration and \( \lambda = 620 \) nm from a colliding-pulse modelocking dye laser generate an electron-hole plasma with a carrier density \( N \approx 3 \times 10^{17} \) cm\(^{-3}\) in the semiconductor inclusions. A weaker probing pulse samples the absorption changes as a function of the delay time between exciting and probing pulses. All data were recorded at room temperature.

Fig.1a,b shows the time-resolved absorption changes for the semiconductor-doped glass filters RG 645 and RG 830. The absorption change \( \Delta \alpha \) is plotted versus delay time between exciting and probing pulses. The dashed curves are the cross-correlation traces determining time zero. In the RG 830 semiconductor-doped glass (Fig.1b) the energy of the femtosecond laser pulses is well above the bandgap, creating carriers with an excess energy \( \Delta E \approx 500 \) meV. A fast recovery of the initial bleaching is observed with a time constant of \( 230 \) fs. This process can be identified with cooling of the initially hot electron gas to the lattice temperature. A comparison with the expected energy loss rate of the electron gas due to the emission of LO-phonons in crystalline semiconductors /3/ yields similar relaxation times for the absorption recovery. In RG 645 (Fig.1a) the opposite extreme is realized. The excess energy is only slightly larger than the bandgap. The absorption decreases with the integrated pulse intensity. No fast carrier relaxation process is observed since the carriers are injected with low excess energy. Consequently, the temperature of the electron gas will not change and no fast absorption recovery is observed. The bleaching is due to the filling of states in the conduction and valence bands and remains essentially constant over the plotted time.

*Present address: AT&T Bell Laboratories, 600 Mountain Ave., Murray Hill, N.J. 07974, USA
Actually, $\Delta A$ decays with a long time constant of $\sim 100$ ps (not shown in the figure). We attribute the 100 ps time constant to electron-hole recombination. This short recombination time is not unreasonable in the light of the large number of surface states in the semiconductor-doped glasses, reacting as efficient recombination centers.

The most interesting case is the semiconductor-doped glass RG 715 (Fig.2), where the initial carrier excess energy $\Delta E$ is $\sim 300$ meV. Here, both the femtosecond decay process as well as the bandfilling can be traced. The three solid curves correspond to different excitation densities, where the lower curves are for two times (curve b) and four times (curve c) attenuated laser pulses, respectively. A semilog plot of the data reveals two time constants; 250 fs for the fast initial absorption recovery and 85 ps for the carrier recombination process. Significant is the superlinear decrease with carrier density of $\Delta A$ for times > 1 ps: $\Delta A$ decreases four times faster if the carrier density is reduced by a factor of 2.

The data can be explained assuming a carrier density dependent bandfilling process. In particular, we think of energy levels or traps below the conduction band, acting as a sink for the conduction band electrons. If the density of these levels is about the same as the carrier density ($\sim 10^{17}$ cm$^{-3}$) and the trapping time is several hundred femtoseconds, the capture of the carriers into these levels becomes saturated, leading to the nonlinear dependence of $\Delta A$ on carrier density observed in Fig.2. The fast 250 fs relaxation process is a combination of cooling of the electron gas due to LO-phonon emission and trapping of charge carriers. From there, electron-hole recombination takes place via surface states in the middle off the bandgap with a time constant of 85-100 ps.
In conclusion, we point out that the absorption changes observed for the three semiconductor-doped glasses investigated here can be well explained within the framework of bulk semiconductor physics. The observed processes are cooling of a hot electron, electron trapping, and recombination. There is no indication of quantum size effects in the currently used glasses. We expect that excitonic effects will only be apparent for semiconductor-doped systems with smaller crystallites and with homogeneous size distribution.

1 G. Bret, F. Gires, Appl. Phys. Lett. 4 (1964) 175
3 J. Shah, Solid State Electron. 21 (1978) 43