SYMPOSIUM U

Ultrafast Nonlinear Optical Phenomena

November 28 – 30, 2000

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SESSION U1:
Chair: Ilias E. Perakis
Tuesday Morning, November 28, 2000
Room 303 (Hynes)

8:30 AM U1.1
NONLINEAR OPTICS IN CHALCOGENIDE GLASS. H.Y. Hwang, G. Lenz, J. Zimmermann, S. Spaelter, T. Katsufuji, S-W. Cheong and R.E. Slusher, Lucent Technologies, Bell Labs, Murray Hill, NJ.

The large demand for higher capacity optical networks has led to the re-examination of the possibility of all-optical processing. We are studying chalcogenide glasses due to their enhanced Kerr nonlinear refractive index n_2, which can be used in a number of applications such as optical switching, photonic bandgap structures, etc. In a bond-orbital model for n_2, three important variables can be identified which contribute to the Kerr nonlinearity: the bandgap, the linear refractive index, and the density of bonds contributing to the band edge. For optimization at communications wavelengths ($\sim$1.55 mm), materials requirements include a bandgap near 1.6 eV, low linear and nonlinear optical attenuation, and a nonresonant n_2 that is 2-3 orders of magnitude above that of silica. We have fabricated and studied a wide range of bulk chalcogenide glasses, and have identified materials with nonlinearities as high as 500 times silica at 1.55 mm. We have also demonstrated record phase shifts ($\sim 2\pi$) by self-phase modulation using $\sim$250 fs pulses in chalcogenide planar waveguides grown by pulsed laser deposition.

8:45 AM *U1.2
ULTRAFAST TERAHERTZ/OPTICAL PROPERTIES OF SEMICONDUCTORS. D.S. Citrin, Washington State University, Dept of Physics and Materials Research Center, Pullman, WA.

In this talk I review my group's recent work on the nonlinear THz/optical properties of semiconductor heterostructures. I discuss coherent mixing phenomena at low optical excitation but in strong THz fields (e.g., THz sideband generation and THz harmonic generation) as well as how strong THz fields can be used to modify the optical properties of highly excited semiconductors (e.g., semiconductor optical amplifiers). Applications for sources and ultrahigh-bandwidth optical switching are discussed.

9:15 AM *U1.3
ULTRAFAST TECHNOLOGY: WHAT IT CAN DO FOR MATERIAL SCIENCE, AND THE REVERSE. Wayne H. Knox, Bell Laboratories Lucent Technologies, Holmdel, NJ.

We discuss some critical recent advances in ultrafast technology that have been enabled by advances in material sciences and reflect on some important new directions in this area. In addition, we examine the reverse situation in which these advances in ultrafast technology can now be put to use in the making of new types of materials. All of these advances are made possible by the unique properties of ultrashort optical pulses.

10:15 AM *U1.4
ANOMALOUS COHERENT BACKSCATTERING OF LIGHT FROM OPAL PHOTONIC CRYSTALS. J. Huang, N. Eradat, M.E. Raikh, Z.V. Vardeny, Dept of Physics, University of Utah, Salt Lake City, UT; A.A. Zakhidov, R.H. Baughman, Honeywell Int., Research and Technology Center, Morristown, NJ.

We studied coherent backscattering (CBS) of light from opal photonic crystals at different incident angles and wavelengths. We observed a dramatic broadening of the CBS cone for incident angle satisfying the Bragg condition. This broadening is explained as due to the light intensity decay in course of propagation along the Bragg direction *before the first* and *after the last* scattering events. Analyzing the cone width as a function of incident angle we were able to extract *both* the Bragg attenuation length and the light mean free path. This shows that CBS measurements are a unique experimental technique to study photonic crystals with disorder when other experimental methods become ambiguous due to disorder-induced broadening.

10:45 AM *U1.5
HIGH ORDER CORRELATIONS IN SEMICONDUCTOR NONLINEAR OPTICAL RESPONSE. Sarah R. Bolton, Physics Department, Williams College, Williamstown, MA; Ulrich Neukirch, Institut fur Festkorperphysik, Univeristat Bremen, Bremen, GEMANY; V.M. Axt, Institut fur Festkorpertheorie, Westfalische Wilhelms Universitat Munster, Munster, GERMANY.

Until recently, the nonlinear optical response of semiconductors has been modeled using the semiconductor Bloch equations. These equations include 2-particle interactions, such as the Coulomb potential which produces the exciton, but neglect 4-particle correlations, of which the bound biexciton is a prominent example. We present here three experiments which measure the role of correlations at the four particle level and beyond. Experiments were performed on a single, 5nm ZnSe quantum well, which combines large exciton and biexciton binding energies with a small exciton linewidth. It thus allows for exclusive excitation of heavy-hole excitons and biexcitons and yields a well resolved spectral response. Semiconductor microcavities have been the subject of intensive investigation as they allow independent control of the two components of the light - matter interaction, i.e. the modes of the electromagnetic field and the electronic states of the active medium. We present here our spectrally-resolved pump-probe measurements, which unambiguously reveal the role of biexcitons in the microcavity optical response. Next, we present pump-probe spectra of the ZnSe quantum well without microcavity, which allow us to quantify the influence of high order correlations in both coherent and incoherent regimes. Finally, we present temporally resolved emission spectra in the six wave mixing geometry. This technique is sensitive to correlations of higher order than those have which been measured in previous experiments, and constitutes the first test for the role of correlations beyond the 4-particle level.

11:15 AM U1.6
ULTRAFAST COULOMB-INDUCED DYNAMICS OF QUANTUM WELL EXCITONS IN MAGNETIC FIELD. T.V. Shahbazyan, N. Primozich and I.E. Perakis, Vanderbilt Univ, Dept of Physics and Astronomy, Nashville, TN.

We study theoretically the ultrafast nonlinear optical response of quantum well excitons in perpendicular magnetic field. We show that, in the coherent regime, the interplay of the biexcitonic and exciton-exciton scattering effects leads to oscillations of the time-integrated four-wave-mixing signal for long negative time delays. We also show that height of the exciton peak in spectrally-resolved signal is modulated with the period determined by the biexciton binding energy. In strong magnetic field, we derive an analytic expression for the memory kernel that takes into account non-perturbatively both biexcitonic and scattering processes. Combining this solution with the exact numetical calculations, we discuss the physical origin of the non-Markovian time evolution of the optical polarization.

11:30 AM *U1.7
ULTRAFAST PHASE TRANSITIONS IN SEMICONDUCTORS. Eric Mazur, Albert M.-T. Kim, Chris A.D. Roeser, J. Paul Callan, Harvard University, Division of Engineering & Applied Sciences and Department of Physics, Cambridge, MA.

We present measurements of the dielectric function of various semiconducting materials (c-GaAs, a-GaAs and GeSb thin films) over a broad energy range (1.5 - 3.5 eV) with a time resolution of 70 fs after the excitation with an ultrashort laser pulse. The time evolution of the dielectric function provides a wealth of information that allows identification and tracking of the electronic and structural dynamics triggered by the pump pulse. At elevated fluence levels all materials undergo a semiconductor to metal transitions.

SESSION U2:
Chair: Daniel S. Chemla
Tuesday Afternoon, November 28, 2000
Room 303 (Hynes)

1:30 PM *U2.1
OPTICAL MANIPULATION OF ELECTRON AND NUCLEAR SPINS IN SEMICONDUCTORS. G. Salis, J.M. Kikkawa, D.T. Fuchs and D.D. Awschalom, Department of Physics, University of California, Santa Barbara, CA.

There is a rapidly growing interest in the use of carrier spin in semiconductor quantum structures as a medium for the manipulation and storage of quantum information. This may eventually form the basis for new paradigms of device operation with improved speed and fundamentally different functionality. Femtosecond-resolved optical experiments reveal a remarkable resistance of quantum spin states to environmental decoherence in a variety of semiconductors and nanostructures, providing hope for the use of these systems as a foundation for the emerging field of spintronics. Optical pulses are used to create a superposition of the basis spin states defined by an applied magnetic field, and to follow the phase, amplitude, and location of the resulting electronic spin precession. The data reveal that spin lifetimes can exceed 100 nanoseconds and that spin packets can be transported 100s of microns. Spatial imaging and dynamical magnetometry of local fields monitors decoherence and dephasing of itinerant spin information as it flows not only through semiconductors and across dissimilar material interfaces, but also into systems of localized moments such as nuclei. Periodic excitation of the electronic spin system can be used to resonantly operate on the nuclear spin

system of the semiconductor host and to detect associated changes in the nuclear magnetization, thereby demonstrating all-optical nuclear magnetic resonance (NMR). These results provide exciting opportunities for transporting and controlling quantum information in the solid state.

2:00 PM *U2.2
MANY-BODY SPIN CORRELATIONS IN NONLINEAR OPTICAL SPECTROSCOPY. Tigran V. Shahbazyan, Dept of Physics and Astronomy, Vanderbilt Univ, Nashville, TN.

We investigate the role of spin correlations in nonlinear absorption due to optical transitions from a deep impurity level to states above a Fermi sea. We demonstrate that the Hubbard repulsion between two electrons occupying the impurity state leads to a logarithmic divergence of the third-order nonlinear optical susceptibility $\chi^{(3)}$ at the absorption threshold. This divergence is a manifestation of the Kondo physics in the nonlinear optical response of Fermi sea systems. We also show that, for off-resonant pump excitation, the pump-probe spectrum exhibits a narrow peak below the linear absorption onset. The light-induced Kondo temperature, which governs the shape of the Kondo-absorption spectrum, can be *tuned* by varying the intensity and frequency of the pump optical field.

2:30 PM *U2.3
NONLINEAR OPTICS OF MOTT INSULATORS. J. Steven Dodge, Matls Sci Div, E.O. Lawrence Berkeley Natl Lab, Berkeley, CA & Physics Dept, Simon Fraser Univ, Burnaby, BC, CANADA.

We have applied time resolved nonlinear optical spectroscopy to probe the electronic structure and nonequilibrium properties of magnetic oxides, the archetypical Mott insulators. Electrical insulation in these materials is driven not by the Pauli exclusion principle, as in conventional semiconductors, but by strong Coulomb repulsion among the electrons of the valence band. Consequently, they are characterized by complex interactions among spin, lattice, orbital and charge degrees of freedom, which are observable in pump-probe experiments as well as in conventional linear spectroscopy. For example, we have studied the spin dynamics of Cr_2O_3 on a picosecond time scale with a near-infrared laser, by exploiting the interaction of spin-waves with spin-flip excitons at much higher energies*. More recently, we have studied the nonequilibrium properties of the charge-transfer gap excitation in $Sr_2CuO_2Cl_2$, a two-dimensional insulating cuprate. In this material, the optical gap exhibits and excitonic peak with a strong temperature-dependence, indicating unusually strong coupling between the exciton and phonons. Optical pumping at the gap energy reveals a pump-probe lineshape which is dominated by this interaction. By relating the amplitude of the pump-probe lineshape to the temperature-dependent linear absorption, we obtain an independent measure of the exciton-phonon coupling. We have also observed optical third harmonic generation in this material, and examined the anisotropy of the third-order susceptibility with polarization analysis. This anisotropy is sensitive to the shape of the chemical bonds in the material, and has been studied in conventional semiconductors but never before in Mott insulators. Further developments from ongoing experiments in this material will be discussed.
*J.S. Dodge, *et al.*, Phys. Rev. Lett. **83**, 4650 (1999).

3:30 PM *U2.4
OBSERVATION OF THERMAL ELECTRON PRESSURE IN THE VIBRATION DYNAMICS OF METAL NANOPARTICLES. G. von Plessen, M. Perner, S. Gresillon, J. Feldmann, University of Munich, Dept of Physics, Munich, GERMANY; J. Porstendorfer, K.-J. Berg, G. Berg, University of Halle-Wittenberg, Dept of Physics, Halle, GERMANY.

Metal nanoparticles exhibit confined acoustic vibrational modes with discrete frequencies. In the time domain, these modes have been observed as periodic signal modulations in ultrafast optical pump-probe experiments on spherical metal nanoparticles [1-3]. The vibrations in these experiments are excited by the thermal expansion of the laser-heated particles, which has been argued to result from the lattice anharmonicity [2,3]. Here we investigate the vibration dynamics of ellipsoidal silver nanoparticles, using time-resolved optical pump-probe spectroscopy. When excited with femtosecond laser pulses, the particles execute anisotropic shape oscillations. An analysis of the time dependence of the vibrations shows that the lattice anharmonicity is not the only mechanism driving the thermal expansion of the particles; there is an additional contribution from the thermal pressure of the optically heated conduction electrons. The hot electrons reach peak temperatures of several thousand Kelvins in these experiments, much higher than the corresponding lattice temperatures. This non-equilibrium situation brings the electron-pressure contribution to the thermal expansion into a range where it becomes comparable to the contribution from the lattice anharmonicity [4].

[1] M. Nisoli et al., Phys. Rev. B 55, 13424 (1997).
[2] J.H. Hodak et al., J. Chem. Phys 108, 9210 (1998).
[3] N. Del Fatti et al., Physica B 263, 54 (1999.
[4] M. Perner et al., Phys. Rev. Lett. (in press).

4:00 PM *U2.5
ULTRAFAST ELECTRON INTERFACE DYNAMICS AND INTERACTIONS IN METAL NANOCRYSTALS. Christos Flytzanis, Fabrice Vallée, Christophe Voisin, Panagiotis Loukakos, Laboratoire d'Optique Quantique, Ecole Polytechnique, Palaiseau, FRANCE.

We summarize the most recent studies of the photoinduced (optical Kerr) nonlinearities in metal nanoparticles in glasses and describe them in terms of a Surface plasmon resonance "Stark shift" mediated through the dielectric confinement and the hot electron mechanism. We exploit the sensitivity of these features to address the evolution of the off-equilibrium electron dynamics and the onset of thermalization using a very sensitive two-color femtosecond pump-probe technique with ~20fs time resolution. We also analyze the electron-phonon coupling and breathing modes of the nanoparticles and the different processes are separated through their specific dependence on the size and form of the metal nanostructures. The impact of pressure and magnetic fields is also analyzed.

4:30 PM *U2.6
DYNAMICS OF RESONANTLY EXCITED CARRIERS IN InAs QUANTUM DOTS. Jagdeep Shah, Lucent Technologies, Bell Laboratories Holmdel, NJ.

The nature of relaxation processes in quantum dots is of considerable current interest. We have performed femtosecond degenerate differential transmission measurements with two special aspects: resonant excitation, severely restricting the processes that contribute to the initial dynamics, and extremely high sensitivity. The magnitude of the initial bleaching allows a determination of the absorption strength of the quantum dots. The dynamics of the bleaching provides new insights into the relaxation processes in quantum dots. In particular, we find unexpectedly high thermal activation rates (~1/ps at 300 K) for carriers photoexcited resonantly in the lowest electronic states of 1.3 micron-wavelength InAs quantum dots. The thermal activation rates have a very strong temperature dependence that can not be explained by single (LO) or two (LO LA) phonon processes. We discuss how thermal activation by multiple LA phonons would explain the strong temperature dependence. These results show that phonon bottleneck does not exist in these quantum dots even in the absence of Auger processes.
This work was performed in collaboration with D. Birkedal, J. Bloch, N. Bonadeo, L.N. Pfeiffer, F. Quochi and K. West.

SESSION U3:
Chair: Gero von Plessen
Wednesday Morning, November 29, 2000
Room 303 (Hynes)

8:30 AM *U3.1
VIBRATIONAL DYNAMICS OF HYDROGEN-DECORATED DEFECTS IN CRYSTALLINE SILICON. Gunter Lüpke, The College of William and Mary, Dept of Applied Science, Williamsburg, VA; Michael Budde, Christine Parks Cheney, Norman Tolk, Len Feldman, Vanderbilt University, Dept of Physics and Astronomy, Nashville, TN.

Vibrational dynamics of isolated hydrogen in crystalline silicon is studied with time-resolved and conventional infrared spectroscopy. Isolated hydrogen at the bond-center site in silicon is formed by proton implantation at cryogenic temperatures. It gives rise to a strong infrared absorption line at 1998 cm^{-1} due to the fundamental excitation of the local vibrational stretch mode, where hydrogen vibrates parallel to the Si-H-Si bond axis. The vibrational lifetime of the first excited state of the stretch mode is measured with pump-probe spectroscopy using the Jefferson Lab. Free-Electron Laser (FEL). The measured lifetime of 7.8±0.2 ps is much shorter than those of Si-H stretch modes on the silicon surface, in amorphous silicon and associated with other hydrogen-related point defects. Measurements in the frequency domain were performed with infrared absorption spectroscopy. The width of the 1998 cm^{-1} line measured at 10 K is found to converge towards its natural line width for decreasing implanted hydrogen concentration. It nearly coincides with the natural line width at the lowest concentrations studied showing that inhomogeneous line broadening is negligible. The temperature dependent line shape of the 1998 cm^{-1} line is in excellent agreement with the theory for vibrational dephasing by energy exchange with two vibrational modes at 112±3 cm^{-1} and 375±8 cm^{-1}, corresponding to perturbed acoustic and optical phonons at the bond-center hydrogen. The lifetimes of the Si-H stretch modes of selected hydrogen-related defects are estimated from their spectral widths and shown to range from 1.6 to more than 37 ps.

9:00 AM *U3.2
SELF GENERATED MAGNETIC CENTRIFUGE FOR SEPARATING ISOTOPIC IONS IN LASER PLASMAS AND ITS USE FOR DEPOSITING ENRICHED THIN FILMS. P.P. Pronko, P.A. VanRompay, Z. Zhang, J.A. Nees, *J.C. Kieffer and G. Mourou Center for Ultrafast Optical Science and Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, MI. *Institute National de la Recherche Scientifique du Quebec, CANADA.

This work investigates the dynamics associated with generation and transport of ultrafast laser plasmas and the isotopic enrichment process that is observed to occur within them. Isotopic separation of ions in these ablation plumes is thought to occur through interaction of the high velocity ions with self-generated magnetic fields occurring as a consequence of hot electron dynamics and non-linear pondermotive forces of the laser light. Methods for modeling and analyzing these phenomena will be presented as well as techniques for directly depositing isotopically enriched thin films.

9:30 AM U3.3
EFFECT OF WAVELENGTH VARIATION ON LASER PROCESSING OF DIELECTRICS BY ULTRASHORT-PULSE, HIGH-REPETITION-RATE LASERS. David R. Ermer, Michael R. Papantonakis and Richard F. Haglund, Jr., Vanderbilt University, Department of Physics and Astronomy, Nashville TN.

Near-infrared femtosecond lasers with MHz pulse-repetition rates are increasingly being used to process dielectric materials, e.g., by etching and drilling. Such a processing cycle has the desirable characteristic that intensity is set by the individual micropulse, while fluence is controlled by the total dose in a macropulse or burst mode. However, at NIR wavelengths (~800 nm or harmonics thereof), electronic excitation leads to excited-state absorption and such undesirable side effects as cracking and crazing. At mid-infrared wavelengths, on the other hand, electronic excitation is negligible, and if the pulse duration is short compared to relevant relaxation times, one can still capture the advantages of ultrashort-pulse materials processing. We used a tunable, mid-infrared, picosecond free-electron laser to ablate both crystalline and amorphous dielectric materials, including silica, calcite and polymers. We find that ablation efficiency depends strongly on the relative magnitudes of the thermal diffusion length and optical absorption depth. The absorption depth can be shortened by orders of magnitude depending on wavelength, and further decreased by nonlinear absorption. In fused silica where absorption increases over five decades from 4 to 9 μm wavelength, a range of behaviors from fracture to explosive vaporization is observed. In crystalline calcite, efficient desorption, ionization and ablation occur at the fundamental ν_2-ν_4 resonance and also at its overtone. Energetic ions and neutrals in the eV range hint at the influence of nonlinear or multiphoton processes, as do measured relaxation times of order 10 ps. Thermal models do not accurately describe the melt depths or ablation yields in either case. A simple explosive vaporization model is more nearly consistent with the experimental results.
Research supported by the Office of Naval Research, Medical Free-Electron Laser Program and by the Office of Science, U.S. Department of Energy.

10:15 AM U3.4
COHERENT VERSUS INCOHERENT DYNAMICS IN InAs QUANTUM DOT ACTIVE WAVEGUIDES. P. Borri, W. Langbein, U. Woggon, Dortmund Univ, Dept of Physics, GERMANY; J.M. Hvam, Denmark Univ, COM Center, DENMARK; D. Bimberg, Berlin Technical Univ, Dept of Physics, GERMANY.

The possibility to optically explore the ultrafast dynamics in quantum dots (QDs), especially by nonlinear experiments such as four-wave mixing (FWM), is often limited by their small interaction volume, resulting in a strongly reduced signal. In this work we present measurements of both time-resolved differential transmission spectroscopy and four-wave mixing in self-organized InAs/InGaAs QDs, using optical waveguiding to enhance the light-matter interaction. A heterodyne detection scheme for co-propagating and co-polarized pump-probe pulses is used. These measurements allow us to investigate the coherent dynamics of strongly confined InAs-based QDs, and the corresponding dephasing mechanisms, and to compare them with the incoherent population dynamics. Samples with different energy separation between the QD ground state and the wetting layer have been measured at room temperature. We find that different confinement energies results in pronounced differences of the population lifetime of the ground-state transition, attributed to thermal activation, with longer lifetimes of several picoseconds for strongly confined QDs. In contrast, the FWM decay is similar in the different samples, showing a photon echo with a dephasing time of $\sim$ 250 fs in the low intensity regime and weakly dependent on the excitation intensity. These results indicate the elastic nature of the dominant dephasing mechanism occurring in the investigated samples, being insensitive to changes in the energy level scheme and not corresponding to a population lifetime. The role of phonon interactions in the measured dephasing is presently under investigation, and temperature dependent measurements are in progress.

10:30 AM *U3.5
SPIN DYNAMICS IN METALLIC FERROMAGNETS. Luca Guidoni, Eric Beaurepaire, Jean-Yves Bigot, IPCMS, Groupe d'Optique Nonlinéaire, UMR 7504 CNRS-ULP, Strasbourg, FRANCE.

One of the challenges in the study of the magnetization dynamics in ferromagnetic materials is to separate, in the magneto-optical response, the contributions associated to the charges and to the spins of the electronic distribution. A few years ago, we have demonstrated that an ultrafast demagnetization can be induced in thin films of nickel and CoPt, by using optical pulses of $\simeq$100 fs duration in a pump probe magneto-optical Kerr configuration. It was shown that the magnetization is modified in the subpicosecond time scale, that is shorter than the electron lattice relaxation time. We have pursued our investigations by performing time resolved ellipsometric measurements with pulses of 20 fs duration in Ni films at saturation under an external magnetic field. With such technique, it is possible to separate the different dynamical components related to the charges and spins of the electrons. In particular, we will show that the nonlinear polarization induced by the ultrashort pump pulse contains a coherent contribution which is proportional to the magnetization but which does not imply a spin dynamics. In addition, the effect of the initial non thermalized electron population on the spin dynamics will be reported. Finally, we will discuss the phase and energy relaxation mechanisms associated to the ultrafast spin dynamics in ferromagnets.

11:00 AM *U3.6
ULTRAFAST OPTICALLY DRIVEN MAGNETIZATION SWITCHING IN THIN FILM MEDIA. Arto Nurmikko, Brown University, Division of Engineering, Providence, RI.

The ultimate speed limits at which magnetism can controlled is of fundamental interest as collective phenomena and of significant application interest in magnetic recording industry. In this presentation a new approach to ultrafast magnetization switching is described, relying on all-optical techniques. We have demonstrated the use of ultrashort pulse laser excitation for optical control of magnetization vector M(t) on a picosecond timescale for a ferromagnetic thin film heterostructures by photomodulating the built-in internal effective magnetic field that arises from the exchange coupling within an antiferromagnetic/ferromagnetic bilayer [1]. The basic idea is to replace a time-varying external magnetic field, employed in conventional magnetization switching, by a time varying internal field, which in this instance arises as a consequence of photoexcitation at the AF/FM interface. That is, we consider the creation of a hot electron/spin gas at or near the heterointerface which breaks up the exchange coupling in the bilayer and abruptly reduces the exchange bias field at a time t=0 corresponding to the arrival of the excitation pulse. This type of selective photoexcitation triggers a dynamical response in the system that leads to coherent rotation of the magnetization vector in the FM layer. We emphasize that, in contrast to magnetization rotation in a time varying external pulsed magnetic field which can be described quite satisfactorily in terms of classical macroscopic variables, we induce here changes in the magnetic energy of the system through the microscopic backdoor, so to speak. As a specific initial material test system, we have chosen the exchange coupled NiFe/NiO (FM/AF) bilayer, characterized by its distinct unidirectional magnetic anisotropy, with important current technological use in giant magnetoresistive and tunnel junction sensors. We have studied systematically the dependence of the magnetization switching and spin relaxation dynamics on the amplitude of exchange bias in the NiFe/NiO system, the former of which can be understood from simple energy arguments and quantitatively described by the Landau-Liftshitz-Gilbert dynamical equations of motion.
In collaboration with G. Ju, L. Chen, R. Farrow, R. Marks, and D. Weller.
Research supported by NSF.
[1] G. Ju et al, Phys. Rev. Lett. 82, 3705, (1999)

11:30 AM *U3.7
MID-INFRARED ULTRAFAST SPECTROSCOPY OF ELECTRON SPIN DYNAMICS IN InAs AND InAs-BASED QUANTUM STRUCTURES. Thomas F. Boggess, J.T. Olesberg, C. Yu, Michael E. Flatté, W.H. Lau, T.C. Hasenberg and E.M. Shaw, University of Iowa, Dept of Physics and Astronomy, Iowa City, IA.

The development of femtosecond optical parametric oscillators (OPO's) operating in the mid-infrared spectral region has led to significant advances in the understanding of ultrafast carrier dynamics

in narrow-band-gap semiconductors. In this work, we use subpicosecond, 3-4 micron wavelength pulses from an OPO to optically inject and probe spin-polarized electron distributions in InAs and InAs/GaSb superlattices. The measured spin dynamics are potentially relevant to the development of new magnetoelectronic devices based on these, and related, materials. Ferromagnetic metals on narrow-band-gap semiconductors, such as InAs, can form low-resistance ohmic contacts that may be suitable for the injection of spin-polarized electrons, a requirement for many proposed magnetoelectronic devices. Furthermore, the InAs/GaSb system is interesting, not only because of its potential application to magnetoelectronic devices, but also because of the unique features influencing spin-relaxation in this system. These superlattices exhibit large conduction band spin splitting that leads to rapid precessional electron spin relaxation at room temperature. Moreover, InAs/GaSb interfaces can incorporate both GaAs and InSb bonds, neither of which occurs within the bulk constituents. The presence of these interface-only bonds, and their different spatial orientations in the interface, contribute to the spin splitting of the band structure. We will discuss temperature dependent measurements of spin relaxation rates in InAs and in InAs/GaSb superlattices grown with controlled interfaces. Measurements will be compared with theoretical results based on a nonperturbative, fourteen-band, restricted-basis-set calculation for the spin-dependent electronic structure.

SESSION U4:
Chair: Tigran V. Shahbazyan
Wednesday Afternoon, November 29, 2000
Room 303 (Hynes)

1:30 PM *U4.1
MULTIDIMENSIONAL FEMTOSECOND CORRELATIONS SPECTROSCOPIES OF ELECTRONIC AND VIBRATIONAL EXCITATIONS. Shaul Mukamel, Andrei Piryatinski, Christoph Scheurer, Vladimir Chernyak, University of Rochester, Department of Chemistry, Rochester, NY.

The response of complex molecules to a series of N femtosecond optical laser pulses provides a multidimensional view into molecular structure as well as a vibrational motions, interactions, and relaxations. In complete analogy with NMR techniques, spreading the spectroscopic information in more than one dimension helps resolve congested spectra, eliminates certain static broadening mechanisms and provides structural and dynamical information unavailable from one-dimensional measurements. The basic concepts underlying these generalized techniques will be surveyed. Femtosecond visible and infrared analogues of multiple-pulse NMR techniques provide novel snapshot probes into the structure and electronic and vibrational dynamics of complex molecular assemblies such as photosynthetic antennae, proteins, and hydrogen-bonded liquids. A classical-oscillator description of these spectroscopies in terms of interacting quasiparticles (rather than transitions among global eigenstates) is developed and sets the stage for designing new pulse sequences and inverting the multidimensional signals to yield molecular structures. Considerable computational advantages and a clear physical insight into the origin of the response and the relevant coherence sizes are provided by a real-space analysis of the underlying coherence-transfer pathways in Liouville space. The multidimensional optical response of the amide I band of glycine dipeptide is calculated using a vibrational-exciton model, treating each peptide bond as a localized anaharmonic vibration. The $2\mathcal{D}$ photon echo signal is obtained by solving the nonlinear exciton equations (NEE). Comparison of different models of spectral broadening (homogeneous and diagonal and off-diagonal static disorder) shows completely different $2\mathcal{D}$ signals even when the $1\mathcal{D}$ infrared spectra are very similar. The phase of the $2\mathcal{D}$ signal may be used to distinguish between overtone and collective types of two-exciton states. Vanishing of the $2\mathcal{D}$ signal along certain directions can be attributed to the variation of the phase. Folding dynamics of β-peptides is studied.
"Multidimensional Femtosecond Correlation Spectroscopies of Electronic and Vibrational Excitations," S. Mukamel, Ann. Rev. Phys. Chem., (1999) (In Press).
"Two-Dimensional Raman Echoes; Femtosecond View of Molecular Structure and Vibrational Coherence", S. Mukamel, A, Piryatinski, and V. Chernyak, *Acct. Chem. Res.* **32**, 145-154 (1999).
"Simulations of Two-Dimensional Femtosecond Infrared Photon-Echoes of Glycine Dipeptide," A. Piryatinski, S. Tretiak, V. Chernyak and S. Mukamel, *J. Raman Spec.*, **31**, 125-135 (2000).

2:00 PM *U4.2
ULTRAFAST MULTIPLE-PULSE PHOTOMODULATION SPECTROSCOPY OF ORGANIC SEMICONDUCTORS.
Sergey Frolov, Bell Laboratories, Lucent Technologies, Murray Hill, NJ.

We study ultrafast relaxation between different excited states in conjugated polymers and other organic semiconductors using a novel ultrafast spectroscopy. The new technique utilizes two excitation pulses and one probe pulse, which are independently tunable across the entire visible and near infrared spectral range. We find that strongly bound excitons are the primary photoexcitations and intramolecular internal conversion is the dominant relaxation mechanism in the organic semiconductors. Due to the large binding energy and fast internal conversion, exciton dissociation with subsequent charge carrier generation is usually an inefficient process. We find that efficient exciton dissociation requires either the assistance of high electric field or optical re-excitation into so-called charge transfer states. We show that the latter states mediate charge separation, so that exciton dissociation occurs even without the applied electric field.

2:30 PM *U4.3
REAL-TIME FEMTOSECOND TIME-SCALE OBSERVATION AND CONTROL OF ATOMIC MOTION AT A METAL SURFACE.
Hrvoje Petek, Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, PA.

Dynamics of the Cs atom motion above the Cu(111) surface following electronic excitation with light are studied with femtosecond time resolution [1,2]. Unusual changes in the surface electronic structure associated with the Cs anti-bonding state within 160 fs after excitation, as observed by time-resolved two-photon photoemission spectroscopy, are attributed to atomic motion in a Cu-Cs bond-breaking process. Describing the change in the energy of the Cs anti-bonding state with a simple classical model provides information on the mechanical forces acting on Cs atoms that are "turned on" by photoexcitation. These forces can lead to alkali atom desorption, or much more probably, local deposition of the 3.1 eV photon energy into phonons and e-h excitations. Coherence in the excitation process is used to selectively generate ground state or excited state nuclear wave packets.
1. S. Ogawa, H. Nagano and H. Petek, Phys. Rev. Lett. 82, 1931 (1999).
2. H. Petek, M.J. Weida, H. Nagano and S. Ogawa, Science 288, 1402 (2000).

3:30 PM *U4.4
THIRD-HARMONIC GENERATION IN TWO-DIMENSIONAL COPPER-OXIDES. A. Schülzgen, P.-A. Blanche, J. Lee, N. Peyghambarian, Optical Sciences Center, University of Arizona, Tucson, AZ; Y. Kawabe, E. Hanamura, A. Yamanaka, H. Sato, Chitose Institute of Science and Technology, Chitose-shi, JAPAN; M. Naito, NTT Basic Research Laboratory, Atsugi-shi, JAPAN; N.T. Dan, Cooperative Excitation Project ERATO, Japan Science and Technology Corporation, Kanagawa, JAPAN; S. Uchida, Y. Tanabe, Department of Applied Physics, University of Tokyo, Tokyo, JAPAN.

Recently, there has been considerable interest in highly correlated electronic systems. A prominent example for such a system is La_2CuO_4 which is also a parent crystal of high temperature superconductors. Not only transport phenomenona such as high-T_c superconductivity, but also the optical response is dominated by the 2-dimensional CuO_2-network. Strong correlation of $3d$-electrons and large overlap between Cu ($3d_{x^2-y^2}$) and O ($2p_\sigma$) orbitals result in large oscillator strength of the charge transfer (CT) excitations. Utilizing tunable femtosecond laser pulses, we demonstrate strong third-harmonic generation (THG) in thin films of La_2CuO_4 between 1.86 eV and 3.21 eV. In contrast to the linear absorption, the THG spectrum consists of sharp structures even at room temperature. This is due to THG enhancement by three-photon resonance with dipole-allowed CT modes and by two-photon resonance with dipole-forbidden CT modes, and, in addition, quantum interference effects between different modes. Taking the linear absorption into account, the THG features are assigned in excellent agreement with respect to both odd and even symmetry CT modes obtained by our excitonic cluster model. Gaining detailed information on the electronic structure of the highly correlated electronic system in La_2CuO_4, our results emphasize the advantages of combining nonlinear THG spectroscopy with conventional linear absorption measurements.

4:00 PM *U4.5
FEMTOSECOND MID-INFRARED STUDIES OF $YBa_2Cu_3O_{7-\delta}$.
T. Elsaesser, R.A. Kaindl, M. Woerner, Max-Born-Institute, Berlin, GERMANY; D.C. Smith, J.F. Ryan, Dept. of Physics, Clarendon Laboratory, Oxford University, Oxford, UNITED KINGDOM; G.A. Farnan, M.P. McCurry, D.G. Walmsley, Dept. of Pure and Applied Physics, Queen's University of Belfast, Belfast, UNITED KINGDOM.

Optical spectra of high-transition temperature superconductors in the mid-infrared have been studied extensively in steady-state experiments. The spectra of both optimally doped and underdoped materials display a gap of in-plane conductivity whose role for superconductivity remains unresolved. Femtosecond studies of the

mid-infrared reflectivity of $YBa_2Cu_3O_{7-\delta}$ after nonequilibrium optical excitation demonstrate the ultrafast fill-in of this gap and reveal two gap constituents: a picosecond recovery of the superconducting condensate in optimally doped and underdoped material and, in the underdoped case, an additional subpicosecond component related to pseudogap correlations. The temperature-dependent amplitudes of both contributions correlate with the antiferromagnetic peak observed at 41 meV in inelastic neutron scattering, supporting the coupling between charges and spin excitations.

4:30 PM *U4.6
OPTICAL-PUMP TERAHERTZ-PROBE STUDIES OF STRONGLY CORRELATED ELECTRON SYSTEMS. Antoinette J. Taylor, Richard D. Averitt, Vanner K. Thorsmølle, Stuart A. Trugman, Materials Science and Technology and Theoretical Divisions, Los Alamos National Lab, Los Alamos, NM.

Optical-pump terahertz-probe spectroscopy is a powerful tool for probing low-energy dynamics in strongly correlated electron systems. We present results of our studies on high-T_c superconductors and mixed valence manganites. For the superconductor $YBa_2Cu_3O_7$, picosecond conductivity measurements probe the interplay between Cooper-pairs and quasiparticles. The conductivity recovery (i.e. reformation of Cooper-pairs) is 1.5 ps at 10K increasing to $\sim$ 4 ps near T_c = 88K. We believe this temperature dependence is related to the anharmonic decay of phonons having energies greater than the superconducting gap. We have also observed a dependence on oxygen doping. Ultrafast picosecond measurements of optically induced changes in the absolute conductivity (0.4-1.0 THz) of $La_{0.7}M_{0.3}MnO_3$ thin films (M = Ca, Sr) from 10K to $\sim 0.9T_c$ reveal a two-component relaxation. A fast, $\sim$2 ps, conductivity decrease arises from optically induced modification of the effective phonon temperature. The slower component, related to spin-lattice relaxation, has a lifetime that increases upon approaching T_c from below in accordance with an increasing spin specific heat. Our results indicate that for $T \ll T_c$, $\partial\sigma/\partial T$ is dominated by incoherent phonons while spin fluctuations dominate near T_c.

SESSION U5: POSTER SESSION
Wednesday Evening, November 29, 2000
8:00 PM
Exhibition Hall D (Hynes)

U5.1
FREE-ELECTRON LASERS: USER FACILITIES FOR ULTRA-FAST STUDIES. Michael J. Kelley, Thomas Jefferson National Accelerator Facility, Newport News, VA and Dept of Applied Science, College of William and Mary, Williamsburg, VA.

Ultra-fast optical phenomena have attracted the attention of many researchers world-wide. An enormous volume of work is at hand to be done. Unfortunately, the high cost of experimental facilities is a barrier to many researchers who might otherwise make critical contributions. However, an alternative to costly laboratory lasers is available and is rapidly expanding. These are the 33 free electron laser (FEL) facilities worldwide, now entering their third generation. User facilities make available average powers to more than 1700 watts with picosecond and shorter pulses over a wide range of wavelengths. The next three years will see a considerable expansion of both capabilities and beamtime at FEL user facilities.

SESSION U6:
Chair: Shaul Mukamel
Thursday Morning, November 30, 2000
Room 303 (Hynes)

8:30 AM U6.1
SUB-10 FS EXCITED STATE DYNAMICS IN CONJUGATED OLIGOMERS STUDIED BY PUMP-PUSH-PROBE EXPERIMENTS. C. Gadermaier, Ist di Matematica e Fisica, Univ di Sassari, ITALY; G. Lanzani, G. Cerullo, M. Zavelani, S. De Silvestri, CEQSE - Dip di Fisica, Politecnico di Milano, ITALY; G Leising, Inst fuer Festkoerperphysik, Techn Univ Graz, AUSTRIA.

On the dawn of the commercialisation of conjugated polymers and oligomers in optoelectronic devices there are still open questions concerning the electronic structure and transitions of these materials. The recent availability of laser pulses with sub-10 fs duration enables direct monitoring of ultrafast electronic and vibronic transitions as well as molecular oscillations. For a wide range of conjugated materials ultrafast pump-probe experiments have revealed the population dynamics of the primary photoexcitations, singlet excitons S1 as well as triplet excitons T1 and positively and negatively charged polarons (doublets) D1 and D1-. A so far rarely adopted modification of pump-probe techniques is pump-push-probe where the primary excited states created via the pump pulse are re-excited with a second (push) pulse into secondary Sn, Tn and D2 - states, which are expected to undergo ultrafast conversion towards their states of origin as well as other primary excitations. The latter case yields particularly valuable information about exciton and polaron formation. The third (probe) pulse reveals the dynamics of these processes via changes in the absorption from the primary excitations or new absorption features from the secondary excitations. The time scale of these processes requires a very high temporal resolution. For the first time we carried out transient pump-push-probe experiments with a temporal resolution below 10 fs on a variety of conjugated oligomers, such as oligophenyls and oligophenylene-vinylenes. By comparing measurements with the modulation upon the pump beam and those with the modulation upon the push beam the dynamics of the secondary photoexcitations as well as oscillations of the excited molecules can be isolated. We present studies of the relaxation dynamics within the same multiplicity as well as the kinetics of cross-over to species with different multiplicity (inter-system crossing between singlets and triplets, exciton dissociation into charged species,), which typically occur within 100 fs.

8:45 AM U6.2
COHERENT PHONON OSCILLATIONS IN QUASI-ONE-DIMENSIONAL ORGANIC MOLECULAR CRYSTALS. T. Hasche, T.W. Canzler, M. Hoffmann, K. Schmidt and K. Leo, Technische Universität Dresden, Institut für Angewandte Photophysik, Dresden, GERMANY; R. Scholz, Technische Universität Chemnitz, Institut für Physik, Chemnitz, GERMANY.

Recently, there has been large interest in organic materials due to very promising device applications, such as OLED. Particularly attractive for applications are organic molecules which form quasi-one-dimensional molecular crystals with strong orbital overlap between neighboring molecules. For free excitons in such systems, first band structure models are proposed (e.g. M. Hoffmann et al, Chem. Phys., in press). However, the relaxation towards self-trapped states is not well understood. Therefore, a detailed investigation of the exciton-phonon interaction is required. Although phonon dynamics have been studied in great detail for inorganic semiconductors and organic molecules in solution, there are only few results in polymers and, particularly, none in quasi-one-dimensional organic molecular crystals.
Here, we study coherent internal and external phonons in the model system MePTCDI (N-N'-dimethylperylene-3,4,9,10-dicarboximide) using room-temperature pump-probe measurements with a temporal resolution of 35 fs. Pump and probe pulses overlap energetically with the lowest absorption band.
The transmission signal reveals a strong oscillatory pattern superimposed on contributions due to vibrational thermalization and the dynamics of exciton populations. The oscillations decay on a time scale of 2 ps. Fourier-transformation delivers lower-energetic contributions between 5 meV and 13 meV, interpreted as rigid movement of the molecules in the crystal unit cell, as well as higher-energetic modes near 27 meV and 70 meV which are assigned to intra-molecular vibrations. Both the internal vibrations and the external phonon modes are excited by Resonant Stimulated Impulsive Raman scattering. Our results are compared to literature data and model calculations based on density functional theory.

9:00 AM U6.3
ULTRAFAST PHONON–MEDIATED OPTICAL NONLINEARITY IN A THIN OLIGOTHIOPHENE FILM. G. Urbasch, H. Giessen, Fachbereich Physik und Wissenschaftliches Zentrum für Materialwissenschaften der Phillipps-Universität Marburg, Marburg, GERMANY; M. Murgia, R. Zamboni, CNR, Istituto di Spettroscopia Molecolare, Bologna, ITALY; R.F. Mahrt, Max-Planck-Institut für Polymerforschung, Mainz, GERMANY.

In recent years a large effort has been spent on the characterisation of the electro–optical properties of π-conjugated oligomers. These materials represent well established modell compounds of both fundamental and technological relevance.
The six-unit oligomer of polythiophene called α-sexithienyl (T6) has been used in organic light–emitting diodes (OLED) and as an active material in promising prototypes of thin film transistors (TFT). Ordered films are available by vaccum sublimation which allows to control the morphology by the deposition parameters.
We have performed femtosecond differential transmission spectroscopy on T6 films at low temperatures covering a broad range of excitation photon energies.
In the probed spectral region from 2.1 eV to 3 eV we find independent of the resonant excitation photon energy a bleaching of the first singlet with a superimposed excited state absorption. The temporal evolution is the same for the whole singlet manifold and independent of the excitation energy in the range from 3.1 eV down to 2.3 eV. For excitation in the transparency region a phonon-mediated optical nonlinearity induced by the strong pump pulse is observed We find an

induced transmission only present during the temporal overlap of the pump and the probe pulse. This signal is observed at the anti-Stokes energetic position of the pump pulse. We attribute this change in transmission to stimulated "inverse" Raman scattering. We furthermore observe simultaneously Raman gain during temporal overlap of the pump and probe pulse for excitation conditions where the Stokes energetic position of the pump pulse is resonant to the intrinsic photoluminescence transition of α-sexithienyl. The phonon associated with the C=C stretching mode is involved in these coherent light matter interactions.

9:15 AM *U6.4
EARLY EVENTS OF PHOTOEXCITATIONS IN ONE-DIMENSIONAL π-ELECTRON SYSTEMS. Guglielmo Lanzani, Politecnico di Milano, Dept. of Physics, Milano, ITALY.

The primary photo-excitations and their relaxation channels, in π-conjugated one-dimensional systems are investigated by pump-probe experiments with sub-10 fs time resolution. Intra-chain vibrational relaxation is identified in the 50 fs time scale e the following evolution via internal conversion directly monitored. The results are discussed within the phenomenology of strongly correlated electron systems, predicting the presence of competing ionic and covalent relaxation channels that leads to geometrically relaxed optically coupled states or symmetry forbidden dark states. First we will briefly discuss the experiment and the technique used for generating 7 fs pulses in the visible range, based on parametric amplification and double-chirped mirror compression. Then examples will be discussed, based on our recent results on conjugated polymers, oligomers and carotenoids. In particular we will present the case of polidyacetylene single chains in benzene solution. The exciting pulses are shorter than the typical vibrational period of the samples, as a consequence they can initiate a collective vibrational coherence within the molecular ensemble, that reflects the coherence in the single molecules. The coherent motion can be started both in the excited state and ground state molecules, the distinction be rather critical. We use positive and negative chirped pulses in order to elucidate the origin of the nuclear coherent motion. The analysis of the frequency content of the transient transmission difference signal reveals the normal modes of the impulsively excited systems. From the overall data we can extract the mode dephasing, related to vibrational energy redistribution.

9:45 AM U6.5
COHERENCE OF EXCITONS IN A CONJUGATED POLYMER: RESONANT RAYLEIGH SCATTERING AND COHERENT CONTROL. Stephen P. Kennedy, Núria Garro, Richard T. Phillips, Cavendish Laboratory, Cambridge, UNITED KINGDOM.

Optically excited excitons in conjugated polymers have several efficient mechanisms for energy relaxation. For example, the strong coupling between vibrational and electronic modes results in relaxation from excited vibrational states which can occur in a few femtoseconds. Also, excitons can migrate within the distribution of conjugation lengths to lower energy sites. Since energy relaxation contributes to phase relaxation, it is not surprising that previous studies of conjugated polymers have been dominated by incoherent photoluminescence, with no observation of the coherent phenomena shown by excitons in GaAs quantum wells. We have found that reducing the energy of incident photons reduces the phase relaxation rate, enabling us to identify, for the first time, *coherent* resonant Rayleigh scattering from poly(p-phenylene vinylene) (PPV). We have performed two-colour upconversion spectroscopy to measure the dynamics of the emission with a time-resolution of $\sim$230fs. We observe an intense resonant component decaying over the first few picoseconds after excitation, leaving spectrally broad photoluminescence that decays exponentially with a lifetime of 500ps. The dynamics of the initial component are strongly dependent on excitation energy, altering from the decay of the excitation pulse to an exponential decay with a lifetime of 450fs for a reduction in excitation energy of 60meV. For the lower excitation energy, the resonant emission dominates the time-integrated emission, its coherence confirmed by the presence of bright and dark speckles in the far-field emission resulting from constructive and destructive interference from phase-coherent excitons in the PPV film. We conclusively prove that there is a coherent polarisation remaining for hundreds of femtoseconds after the excitation pulse by interfering constructively and destructively with a second pulse delayed with an accuracy of 50 attoseconds ($\sim\lambda/40$). This is the first direct evidence of the control of quantum phase in the excitations of a conjugated polymer.

10:30 AM *U6.6
"SIMULATIONS OF NONLINEAR OPTICAL RESPONSE IN ATOMS, MOLECULES AND SOLIDS". S.T. Pantelides, M. Ferconi, J.J. Vicente Alvarez, Vanderbilt University, Nashville, TN.

Density functional theory has been the method of choice to study ground state properties of solids and, more recently, molecules. Time-dependent density functional theory is well-suited to describe interactions of light with matter in both the linear and fully nonlinear regime. We report initial implementations of the theory. In atoms we show that intense infrared light produces high-harmonic response, including x-rays, as observed experimentally. In molecules we find fascinating third-harmonic response response in the uv. Intense infrared can selectively break bonds at frequencies that are not related to the ground-state normal modes. Initial application to crystalline Si finds odd-harmonic response as expected. This work was supported in part by the Office of Naval Research.

11:00 AM *U6.7
EXPERIMENTS ON QUANTUM KINETICS IN SEMICONDUCTORS. Martin Wegener, Institut fuer Angewandte Physik, Universitaet Karlsruhe, Karlsruhe, GERMANY.

I review recent experiments on quantum kinetics in semiconductors. Quantum kinetics addresses that temporal regime in which the finite duration of the interaction process becomes important. This timescale is usually shorter than the known scattering time, the mean time between collisions. Results on electron-phonon and on electron-plasmon interaction are presented. The latter experiments show a carrier-density dependent oscillation which reflects the transfer of excitation back and forth between individual electrons and the collective of electrons, the plasmon. If time permits, I will also address ongoing experiments with 5fs optical pulses beyond the rotating wave approximation.

11:30 AM *U6.8
QUANTUM KINETICS FOR FEMTOSECOND SEMICONDUCTOR DTS AND FWM SPECTROSCOPY. Hartmut Haug, Institut f. Theoret. Physik, J.W. Goethe-Uni.-Frankfurt, GERMANY.

Femtosecond spectroscopy allows to observe the dynamics of photo-excited carriers on a time scale which is shorter than characteristic periods e.g. of a longituninal optical (LO) phonon or plasmon oscillation, or of an exciton orbit. Under these conditions the relaxation and dephasing kinetics can no longer be described by the semiclassical Boltzmann kinetics. One has to use instead quantum kinetics with its pronounced memory effects due to the quantum coherence of the carriers shortly after their excitation by coherent laser pulses. At higher excitation densities the buildup of screening by the carriers has to be included self-consistently. I will use this quantum kinetic theory together with semiconductor Bloch equations in order to review the analysis of several recent resonant femtosecond differential transmission (DTS) and four-wave mixing (FWM) experiments, also with coherent control. It will be shown that quantum kinetics allows to describe the dephasing and relaxation kinetics due to carrier-LO-phonon and carrier-carrier scattering quantitatively. Quantum coherence in the form of oscillations superimposed on the decaying time-integrated FWM signal due to LO-phonon-plasmon mixed modes will be predicted. These oscillations have recently been observed by M. Wegener et al. in agreement with the theory using resonant 13 fs photon-echo measurements with coherent control in the form of a phase-locked double pump pulse.

SESSION U7:
Chair: Sarah R. Bolton
Thursday Afternoon, November 30, 2000
Room 303 (Hynes)

1:30 PM *U7.1
COHERENT EFFECTS IN SEMICONDUCTOR LIGHT EMISSION. S.W. Koch, M. Kira, F. Jahnke and W. Hoyer, Dept. of Physics, Univ. of Marburg, Marburg, GERMANY.

Coherent signatures in the secondary emission from short pulse excited semiconductor quantum wells are discussed. Signatures of excitation induced dephasing, coherent control and non-classical light statistics are analyzed.
Reference:
M. Kira et al., Prog. Quant. Electron. 23, 189 (1999).

2:00 PM *U7.2
WHAT CAN WE LEARN FROM RESONANT RAYLEIGH SCATTERING OF EXCITONS? R. Zimmermann, E. Runge, V. Savona, Inst of Physics, Humboldt Univ Berlin, GERMANY.

After optical excitation of a semiconductor nanostructure, a wealth of information is contained in the subsequent secondary light emission. It comprises Rayleigh scattering (coherent with the driving field) and luminescence (incoherent). Rayleigh scattering is due to disorder in the sample which localizes the exciton motion, and the time- and angle-resolved signal is simulated on the basis of disorder eigenstates. This provides insight into quantum mechanical features of the exciton states as localization lengths, wave function statistics, and level

repulsion. Among several experimental set-ups, we emphasize enhanced resonant backscattering. The dependence on the underlying disorder allows to draw conclusions on the interface quality in the nanostructure.

2:30 PM U7.3
QUANTUM KINETIC DESCRIPTION OF PHONON-INDUCED CARRIER CAPTURE. V.M. Axt, M. Glanemann, M. Herbst, T. Kuhn, Institut für Festkörpertheorie, Univ. Münster, GERMANY.

Quite generally, on ultrafast time-scales scattering events show signatures of their finite duration and thus require a quantum kinetic description which naturally includes energy-time uncertainty. Analogously, on ultrashort length-scales the Boltzmann picture of scattering events taking place at a certain position between well-defined momentum states looses its validity due to the momentum-position uncertainty. We present a quantum kinetic analysis of the ultrafast carrier dynamics in spatially inhomogeneous systems based on the density matrix formalism. We apply the formalism to the case of an electronic wavepacket travelling across a potential well where carriers can be trapped in the well by phonon emission. In contrast to a semiclassical (Markov) description the quantum kinetic formulation is independent of the choice of the single-particle basis. This is particularly advantageous when, as in the case of carrier trapping, the scattering takes place between states of different dimensionality. The system under investigation is a quantum dot embedded in a one-dimensional quantum wire. We study the dynamics of wavepackets prepared at a certain distance from the dot with kinetic energies below the threshold for optical phonon emission. When the wavepacket approaches the well part of the wavepacket is trapped, the remaining part being transmitted or reflected. The separation of the incoming wavepacket into these three parts exhibits a complicated time dependence as long as they spatially overlap and interfere. By comparing the dynamics including phonon interaction with a free wavepacket scattered only by the potential well, we clearly distinguish the capture process from reflection and transmission. Without phonons the mean kinetic energy increases while the wavepacket is above the well and falls back to its initial value after passing the well. With phonons the final value differs from the initial value and the time dependence of the kinetic energy reveals many details of the capture process.

3:15 PM *U7.4
SATURATION OF NORMAL-MODE COUPLING IN LATERALLY CONFINED NANOCAVITIES - TOWARD THE QUANTUM STATISTICAL LIMIT. Ch. Spiegelberg, H.M. Gibbs, E.S. Lee, C. Ell, P. Brick, G. Khitrova, Optical Sciences Center, University of Arizona, Tucson, AZ; D.G. Deppe and D.L. Huffaker, Department of Electrical and Computer Engineering, Microelectronics Research Center, The University of Texas at Austin, Austin, TX.

We present our recent results on normal-mode coupling of excitons and photons in oxide-apertured semiconductor nanocavities. The cavity mode diameters range from 7 to 1 μm. Linear transmission data for different aperture sizes reveal the three-dimensional confinement of the light field. We observe quantized cavity modes which are energetically well separated. The nanocavities show pronounced normal mode coupling with a splitting-to-linewidth ratio of 4.9 for an aperture size of 2 μm. We have determined the number of photons required for saturation of the normal-mode coupling by femtosecond pump-probe experiments. Compared to planar microcavities with no lateral confinement using a 50-μm diameter spot, this number reduces drastically from 2×10^5 photons to about 300 for a cavity with 2 μm mode diameter. Together with a cavity quality factor of 2000 this value is clearly a step forward toward the quantum statistical limit. Eventually, for sufficiently small mode volumes, new quantum effects are expected and quantum entanglement should be possible.

3:45 PM *U7.5
COHERENT OSCILLATORY NONLINEAR OPTICAL RESPONSE FROM INTERACTING EXCITONS AND EXCITON- POLARITONS. Hailin Wang, University of Oregon, Department of Physics, Eugene, OR.

Simultaneous excitation of multiple excitonic transitions by a short laser pulse can lead to coherent oscillatory behaviors in nonlinear optical processes such as transient four wave mixing. These oscillations, while loosely referred to as quantum beats, are strongly influenced by manybody Coulomb interactions inherent in an excitonic system and in many aspects differ qualitatively from quantum beats in atomic systems. In this paper, I will discuss experimental studies of coherent oscillations in transient four wave mixing from a system of heavy and light hole excitons in a GaAs quantum well and a system of exciton-polaritons in a semiconductor microcavity. In both systems, exciton-exciton interactions can induce coherent oscillations that otherwise would be absent in a non-interacting system. For the exciton-polariton system, the relative phase of the coherent oscillation reflect the details of the exciton-exciton interactions, thus providing a fingerprint for the underlying manybody interactions. Theoretical analysis based on modified optical Bloch equations will also be discussed.

4:15 PM U7.6
HUGE ANGLE-RESONANT STIMULATED POLARITON SCATTERING IN SEMICONDUCTOR MICROCAVITIES. Pavlos Savvidis, Jeremy Baumberg, Dept of Physics, Univ of Southampton, UNITED KINGDOM; Mark Stevenson, Maurice Skolnick, Dept of Physics, Univ of Sheffield, UNITED KINGDOM; David Whitakker, Toshiba Research Cambridge Lab, Cambridge, UNITED KINGDOM; John Roberts, Dept of Electronic Engineering, Univ of Sheffield, UNITED KINGDOM.

With the advent of highly controlled semiconductor epitaxies, both electronic and photonic wavefunctions and their interaction can be specifically manipulated. We demonstrate here that by devising new dispersion relations for quasiparticles which mix photons and excitons, entirely new dynamic scattering properties can be evoked. Semiconductor microcavities are two-dimensional structures in which the excitons confined in a quantum well interact with photons that are confined in a cavity formed by two high reflectivity Bragg mirrors. Coupling of these light and matter modes occurs when their wavefunctions spatially overlap and are matched in both energy and in-plane momentum. This leads to an anti-crossing behaviour producing two new mixed modes known as exciton-polaritons. Recently there has been much discussion over the behaviour of polaritons, and to what extent they differ significantly from excitons. Here we probe the angular properties of microcavities, which directly probes the dispersion relation of these quasi-particles. Using a femtosecond goniometer, multicolour ultrafast time-resolved reflection, emission and gain measurements are used to track polariton-polariton scattering processes [1,2]. New parametric phenomena are observed as a result of the bosonic nature of the polaritons [2]. Strong amplification of a probe beam dominates at a critical pump angle, due to stimulated scattering obeying energy and momentum conservation on the dispersion relation. As the polariton dispersion relation is modified by changing the cavity thickness, lasing emission in a cone can be observed, with a peculiar dynamics that we can directly observe. This allows us to determine the joint effect of localised and extended exciton states which exist in the confined heterostructures. These scattering processes, imposed by the deformed dispersion relations of polaritons, can be tailored to significantly enhance non-linear optoelectronic properties.
[1] J.J. Baumberg et al., Phys. Rev. Lett. 81, 661 (1998).
[2] P.G. Savvidis et al., Phys. Rev. Lett. 84, 1547 (2000).

4:30 PM U7.7
ULTRAFAST DYNAMICS OF A POLARITON–BI-EXCITON TRANSITION IN A III-V SEMICONDUCTOR MICROCAVITY. P. Borri, W. Langbein, U. Woggon, Dortmund Univ, Dept of Physics, GERMANY; J.R. Jensen, J.M. Hvam, Denmark Univ, COM Center, DENMARK.

In this work, we have investigated the nonlinear dynamics of a high-quality quantum well (QW) microcavity. The sample consists of a 25 nm GaAs single QW in the center of a wedged λ-cavity with AlAs/AlGaAs Bragg reflectors. The very high quality of the sample is evidenced by a ratio between the heavy-hole (HH) Rabi splitting and the polariton linewidth as large as 19 near zero detuning. The HH Rabi splitting is approximately four times larger than the biexciton binding energy in the bare quantum well. Due to the narrow polariton linewidths a spectrally well-resolved nonlinear optical absorption can be observed, associated to the transition from the lower polariton to the biexciton. Time-resolved pump-probe spectroscopy of this new transition at low temperature (5 K) is performed with subpicosecond resolution. The dependence from the polarization of the exciting pulses, from their relative delay time, and from the pump excitation intensity clearly reveals the biexcitonic nature of the measured transition and its dynamics related to the lifetime of the lower polariton density. Moreover, measurements at different detunings between the cavity mode and the HH exciton demonstrate that the biexciton binding energy is not significantly affected by the strong-exciton photon coupling in the microcavity. This result is explained when considering that the exciton-photon coupling strongly modifies only a very small region of the in-plane wave vectors in the microcavity, and the constituent excitons in the biexciton extend over a region of large wavevectors due to the biexciton binding. Moreover, in spite of the strong exciton-photon coupling, no signature of bipolariton formation is detected, consistent with expectation in theory due to the small effective mass in the lower polaritons. Additional four-wave mixing measurements are in progress.