FINAL REPORT

SUBMITTED TO: National Aeronautics and Space Administration
Langley Research Center
Hampton, Virginia 23681

CONTRACT MONITOR: James Barnes
NASA LaRC, MS 474
Hampton, Virginia 23665

INSTITUTION: Department of Physics
Boston College
Chestnut Hill, Massachusetts 02467

TITLE OF RESEARCH: Nato Advanced Study Institute on Spectroscopy

NASA GRANT NUMBER: NAG-1-01-067

PERIOD COVERED BY THIS GRANT: April 18, 2001 - August 17, 2001

PRINCIPAL INVESTIGATOR: Baldassare Di Bartolo
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PREFACE


This meeting was organized and directed by me, Principal Investigator for the Grant, and co-sponsored by NASA under Grant NAG-1-01-067.

I gratefully acknowledge the NASA sponsorship and the cordial assistance received by the Contract Monitor Dr. James Barnes, Head of the Laser Systems Branch of NASA.

Baldassare Di Bartolo
Professor of Physics
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This booklet presents an account of the course "Spectroscopy of Systems with Spatially Confined Structures" held in Erice-Sicily, Italy, from June 15 to June 30, 2001. This meeting was organized by the International School of Atomic and Molecular Spectroscopy of the "Ettore Majorana" Centre for Scientific Culture.

The purpose of this course was to present and discuss nanometer-scale physics, a rapidly progressing field. The top-down approach of semiconductor technology will soon meet the scales of the bottom-up approaches of supramolecular chemistry and of spatially localized excitations in ionic crystals. This course dealt with the fabrication, measurement and understanding of the relevant structures and brought together the scientific communities responsible for these development. The advances in this area of physics have already led to applications in optoelectronics and will likely lead to many more. The subjects of the course included spatially resolved structures such as quantum wells, quantum wires and quantum dots, single atoms and molecules, clusters, fractal systems, and the development of related techniques like near-field spectroscopy and confocal microscopy to study such systems. The meeting also revisited the well-established topics of luminescence. Each lecturer developed a coherent section of the program starting at a somewhat fundamental level and ultimately reaching the frontier of knowledge in the field in a systematic and didactic fashion. Many participants also presented their current research work in the formats of short seminars and posters.

The participants came from 16 different countries (France, United States, Germany, England, Italy, Switzerland, Russia, Rep. Of Georgia, Belarus, Latvia, Estonia, Finland, Turkey, Portugal, Spain, and The Netherlands).

There were 20 formal lectures and 4 long seminars. In addition, 13 short seminars and 10 posters were presented. Two round-table discussions were held. The first round-table discussion took place on the third day of lectures in order to evaluate the work done in the first few days of the course and consider suggestions and proposals regarding the organization, format and presentation of the lectures. The second one was held at the conclusion of the course, so that the participants could comment on the work done during the entire meeting and discuss various proposals for the next course of the International School of Atomic and Molecular Spectroscopy.

A summary of the lectures, long seminars, short seminars, posters, round-table discussions, etc. is presented here in this booklet. The full-text lectures/long seminars and the abstracts of short seminars and posters will be published as a book by Kluwer Academic Publishers.

The secretary of the course was Matthew Vannette.

I wish to express my sincere gratitude to Mr. Alessandro Noto, Mr. Pino Aceto, Ms. Fiorella Ruggiu, and to all the personnel of the "Ettore Majorana" Centre for their kind assistance. I also wish to acknowledge the sponsorship of the meeting by NATO, NASA, the ENEA Organization, Boston College, the Italian Ministry of University and Scientific Research and Technology, the USA National Science Foundation, and the Sicilian Regional Government.
I would like to thank the Co-Director of the Course, Academian Alexander Voitovich, members of the organizing committee (Prof. Martin Wegener, Dr. Giuseppe Baldacchini, Prof. Claus Klingshirn, Dr. Cees Ronda, Dr. Eric Mazur, Dr. James Barnes, Dr. Norman Barnes, and Prof. Ralph von Baltz), the secretary of the course (Mr. Matthew Vannette), Prof. Xuesheng Chen, and Prof. John Di Bartolo for their helps in organizing the course. I would also like to thank Jennifer Vannette and Monica Cimino and Yueli Chen for their help and Dr. Elisabeth Kurtz for her beautiful drawings, which are graceful additions to these proceedings.

I am looking forward to our activities at the Majorana Centre in years to come, including the next 2003 meeting of the International School of Atomic and Molecular Spectroscopy.

Baldassare (Rino) Di Bartolo
Director of the International School of Atomic and Molecular Spectroscopy of the "Ettore Majorana" Center
Advanced Study Institutes Held
at the "Ettore Majorana" Centre in Erice, Sicily, Italy

1974 – Optical Properties of Ions in Solids
1975 – The Spectroscopy of the Excited State
1977 – Luminescence of Inorganic Solids
1979 – Radiationless Processes
1981 – Collective Excitations in Solids
1983 – Energy Transfer Processes in Condensed Matter
1985 – Spectroscopy of Solid-State Laser Type Materials
1987 – Disordered Solids: Structures and Processes
1989 – Advances in Nonradiative Processes
1991 – Optical Properties of Excited State in Solids
1993 – Nonlinear spectroscopy of Solids: Advances and Applications
1995 – Spectroscopy and Dynamics of Collective Excitations in Solids
1996 – Workshop on Luminescence Spectroscopy
1997 – Ultrafast Dynamics of Quantum Systems: Physical Processes and Spectroscopic Techniques
1998 – Workshop on Advances in Solid State Luminescence Spectroscopy
1999 – Advances in Energy Transfer Processes
2000 – Workshop on Advanced Topics in Luminescence Spectroscopy
2001 – Spectroscopy of Systems with Spatially Confined Structures
Drawings, by Dr. Elisabeth Kurtz
(The first five drawings are scenes in Erice, the sixth and seventh drawings are views of the San Vito beach, the eighth is a view of a temple in Selinunte, and the last drawing represents the temple of Segesta.)
COSTRETTO

Confined Structures in Erice Two Thousand One

Black clouds loom over the historic mount of Erice and the cold penetrates inside the bones like ice, while serious scholars sit motionless on the benches, in a cubic space once the most beautiful among the churches.

San Domenico has been for centuries the Saint Patron but now the hall to Dirac is dedicated for the positron, and noted scientists celebrate the feats of Science while old and newcomers assist in religious silence.

They arrived from several countries far away, like America, China, Russia, Finland and Norway, and from Turkey, Portugal, Estonia, England with France, Germany, Italy and the small Switzerland, and also Spain, Latvia, Georgia and The Netherlands.*

They sleep in secluded franciscan accommodations and eat spartan meals during the intermissions, so that in a while they will become mostly ethereal starting to see phantoms where the objects are only real.

If life in Erice will continue the same way in a little while everything will fade away, and Science will be among the many recollections as the callipygian Venus and her longed donations.

However, the few people loving Science are well known for their stoic endurance, and so, also with the help of the Marsala room, they will remain faithful like a new groom.

Moreover, there have been excursions and banquets that for this School have been always the basic tenets, where good wines flows as rivers on the tables and food is rich and plenty like in the fables.

*The High number of nations broke the symmetry of four verses, but the five-fold degeneracy has been eliminated by the poetic licence which produced a verse band gap.
Thus, they still proceed to the various sessions where they learn with deep appreciations that light is not free to move in the space, especially if photonic band gaps are in place.

However, they are confounded by the information that the MAFIA code solves more than one equation, and the bow-tie is not an elegant piece of the men dress but a semiconductor defect almost dimensionless.

Once to oppose a force it has been added a spring which nobody understood how it became a string, and the audience looses the common sense of balance when quantum beats are mimicked with a Russian dance.

But the information which is more distressing is the tale of teleportation and light stopping, so we do not know whether applying the laws of Physics or better the well known Aristotleian Metaphysics.

Moreover, also the glasses are in search of a definition while once it was enough transparency and insulation, and it is very strange that for a social communication we need an elegant but rather solitary propagation.

Discussions start and die as the stars in the firmament but the spaghetti invention is still a live argument, because Marco Polo has mixed up with a French the story so that the Chinese people still claim a fake victory.

No debate instead even started for the ice cream which Italy tasted while China could only dream, and the American do not like the bare evidence that the genial Columbus invented their own existence.

But also when the controversies reaches high pitches everybody likes to go to the fabulous beaches, where they swim, tan and also try sleeping, though a few risked to be buried by somebody kidding.

Anyway, the School is closing with satisfaction and this is a good omen for its continuation, so I wish it a long life in the third Millennium and all of us to meet again here as the best premium.

Giuseppe Baldacchini, Erice, June 29, 2001
LECTURES

The Role of Structure Confinement in the Spectroscopy of Physical Systems
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These lectures provided the background for many of the concepts that were presented and developed during the course:
1. Review of some quantum classics: particle in a box, harmonic oscillator, central potential, one electron atom
2. Barrier tunneling and electron traps
3. Identical particles. Many-electron atoms. Identical bosons
4. Energy levels of a crystalline solid. Insulators, semiconductors, conductors
5. Superlattices and Nanostructures.

Photonic Structures: Atoms, Molecules & Crystals
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The modification of electromagnetic modes in microcavities and periodic structures on the optical properties is of great interest for both their basic properties and for their potential applications in solid state devices. In structures with sizes on the scale of the wavelength of light, the interaction between light and matter is modified so that the optical properties can change considerably, i.e. the creation of a photonic band gap strongly suppresses spontaneous emission.

The following overview on the theory and experiment of confined electromagnetic fields was presented:
1. Discussion of electromagnetic fields and Maxwell equations:
Waves, modes, and photons. Calculation and measurement of fields.
2. "Photonic atoms":
Metallic and dielectric resonators and their analogy with a LC circuit, particle in a box, and an electronic quantum dot (QD).
3. "Photonic molecules":
Coupled resonators and their analogy with coupled LC circuits, chemical bond (H₂, H₂⁺ - molecules), and coupled electronic QDs.
4. "Photonic crystals":
Periodic dielectric structures and their analogy with a LC chain and electronic crystals.
Bloch waves, band structure, energy transport velocity.
5. Design of photonic structures and some experimental results.
Optical Near-Field Spectroscopy
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Seeing things, i.e. looking at them with our human eye to find out about their color or their surface topography, is one of the most intuitive ways to experience the world around us. Conventional optical microscopes can help us a lot in improving the spatial resolution with respect to the bare eye. But there are limits to the resolution. In school, I learned that the wavelength of light sets these limits. Today, we know that this statement is incorrect in the above form. Indeed, it is correct for conventional, i.e. optical far-field, microscopes. However, this limitation can be overcome by so-called optical near-field microscopes. In this case, ideally, the resolution is only limited by the 'size' of an optical probe. In this lecture I explained the underlying principles on a very basic level and illustrated the capabilities by selected examples from semiconductor physics. This includes spectroscopy and microscopy of nanostructures ('quantum wires', single defects) and disorder potentials (related to 'self-organized quantum dots' and localization) as well as time-resolved transport on a 100 fs- 100 nm scale.

The outline of these lectures follows:

1. Introduction
2. Far-field optical microscopy
   Basics. Scanning confocal microscopy
3. Scanning near-field optical microscopy (SNOM)
   Basics. Different modes of operation: Overview. Aperture SNOM. Depolarization
   SNOM. Apertureless SNOM
4. Examples I: Semiconductor nanostructures
   Quantum wires. Single bow-tie defects in a single semiconductor quantum well
5. Examples II: Disordered semiconductors
   Basics: Random-matrix-theory. Statistical analysis of energy levels
6. Examples III: Transport on a 100 fs - 100 nm scale
7. Summary.

The Future of Laser Technology for Spectroscopy
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The laser is one of the most fantastic and versatile tools invented recently, although its story began with the luminescent phenomena, which have been always at the center of curiosity of mankind since ancient times. However, the important aspects of luminescence were not discovered until the 19th century and only in the 20th century has the various basic and technical knowledge coalesced for the realization of the first laser device. Since then, the field exploded almost exponentially, and thousands of different materials, in the state of solids, liquids, vapors, gases, plasmas, and elementary particles have lased up to now. However, only a few of them became practical lasers, which have been used with outstanding results both in basic science and in industrial and commercial applications. As for the spectroscopic applications,
the laser light has started an unprecedented revolution because of its unique properties of monochromaticity, continuous and pulsed regimes, unrivaled by any other natural and artificial light source.

Due to the enormity of the laser field, only the most important aspects of lasers were considered, starting from the essential historical background, describing the rapid evolution of the main laser systems in the first decades after their invention, the more paced growth in the last decades, and forecasting the near future developments. In general, although a few sectors show signs of maturity, the field as a whole looks alive and well. We may expect a new golden age as a consequence of the impressive technological advances.

Creation, Characterization, and Applications of Ultrashort Laser Pulser

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The following topics were to have been covered:
1. Linear and nonlinear propagation of optical pulses
Interaction of matter with light, dielectric function, response of bound and free electrons, plasmas, pulse dispersion, nonlinear optics, second-harmonic generation, continuum generation, self focusing

2. Femtosecond measurement techniques
Pump-probe technique, dispersion compensation, grating and prism compressors, time and frequency representations of short pulses, temporal characterization of short pulses, joint time-frequency measurements, frequency-resolved optical gating, resolution limits

3. Ultrafast phase transitions in semiconductors
Interaction of light with semiconductors, time scales of relevant processes, from structure to optical properties, broadband time-resolved ellipsometry technique, results for selective semiconductors

4. Microexplosions in transparent dielectrics
Femtosecond laser-induced microexplosions, conditions at focus, morphology of structural changes, energy deposition studies, laser-field ionization and impact ionization, dark-field scattering technique, dynamics microexplosions, applications
Growth and Spectroscopy of Quantum Wires and Quantum Dots

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Quantum wire (QWR) and quantum dot (QD) nanostructures have been in the forefront of research in solid state physics and optoelectronics for more than a decade because of the opportunities they offer for studying low-dimensional electronic systems and exploring novel device applications. To a large extent, this field has been driven by the experimental techniques that have been developed for preparing such systems with controlled potential distribution and non-compromised interface quality. In this lecture we reviewed the different approaches for making such QWRs and QDs, and then concentrated on one of the more successful techniques explored, namely, epitaxial growth on nonplanar substrates. Analytic and numerical models developed for getting an insight into the growth mechanisms on nonplanar substrates were presented and compared with experimental results. In particular, the structure of QWRs, QDs and their superlattice, prepared with this approach using lattice matched or strained compound semiconductors, was described in some detail. It was demonstrated that quantum nanostructures with high interface quality and uniformity, in addition to reasonable control of their potential distribution, can be obtained in this way.

The second part of the lecture was devoted to optical and electrical spectroscopy of these quantum nanostructures. QWRs and QDs grown on nonplanar substrates exhibit efficient luminescence that permits the study of their electronic structure using photoluminescence (PL) and PL excitation spectroscopy. The determination of the one-dimensional (1D) subband separation in the wires and the 0D states in the dots by means of controlling their growth parameters was demonstrated. The effect of structural disorder on the optical spectra of V-groove QWRs, and the impact of Coulomb correlation on the PL spectra of QWRs and QDs was also discussed. In particular, it was shown that excitonic effects dominate the emission and absorption in 1D wires, and that the spectra of QDs are extremely sensitive to the charged impurity distribution in their environment. The effect of photon confinement on the emission spectra of QWRs embedded in 2D, 1D or 0D optical cavities was also described. Electrical conductance measurements in 1D V-groove QWRs was presented and discussed. It is found that quantized electrical conductance with values different than the canonical one (G0=2e2/h) is exhibited by V-groove QWRs, as well as by other wires with a rigid, heterostructure lateral confinement (e.g., T-shaped wires). It was shown that this is due to electron scattering that takes place at the 2D/1D interface between the wires and the electron reservoirs. The impact of intersubband scattering on the conductance of two, serially-connected QWRs was also presented.

The final part was concerned optoelectronic device applications of QWRs and QDs. Applications of these nanostructures in light emitting diodes, diode lasers, waveguide modulators and switches and tips for scanning near field optical microscopy was reviewed.
Theory of the Optical Properties of Quantum Wells, Wires and Dots
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The following topics were presented:
1. Electronic states in confined mesoscopic microstructures.
2. Optical transitions, semiconductor Bloch equations free-particle and excitonic linear spectra.
3. Plasma density-dependent spectra, electro- and magneto-optical spectra.

Bloch Oscillation in Semiconductor Superlattices
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More than 60 years ago, Zener predicted that an electron in a periodic potential subject to a static electric field will perform harmonic “Bloch oscillations” in real space. We presented the results which directly prove the existence of these spatial oscillations: By observing the field-induced shift of the Wannier-Stark ladder, the macroscopic dipole moment associated with Bloch oscillations of the electron wave packets can be traced. The displacement of the electron wave packet can then be derived as a function of time, with the excitation density as the sole parameter.

We discussed the dependence of Bloch oscillation dynamics on the optical excitation conditions. In particular, we showed that the motion can be continuously tuned between the harmonic spatial motion envisioned by Zener and a symmetric breathing-mode motion where no macroscopic dipole oscillation is present. The results further show that excitonic effects due to the photo-excited holes can significantly influence the dynamics of the wave packets.

Optical Anisotropy of Semiconductor Nanostructures
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The optical anisotropy (OA) phenomena and the related anisotropic optical effects in the low-dimensional semiconductor structures were considered.

First the general concepts concerning the OA phenomena and some examples of the anisotropic optical effects in bulk crystals were given. Then the phenomenological description of the OA followed. Some possible reasons which lead to the intrinsic OA in the low-dimensional semiconductor structures were given. Confinement and crystallographic orientation effects on the anisotropy of the optical properties in the quantum wells (one-dimensional confinement) and the quantum wires (two-dimensional confinement) proved to be quite important. In addition, the intrinsic
strain which is generated during growth, leads to the OA. Except for the structures above, the zinc-blend semiconductor alloys that exhibit spontaneous CuPt ordering show the strain-induced intrinsic OA.

The quantum-confined Stark effect in the quantum wells was also discussed as an example of the extrinsic OA. The anisotropy in this case is due to the external electric field. In the case of three-dimensional confinement, i.e., for the quantum dots, we considered the shape and structural anisotropy effects on the OA. Then we discussed the relation between the optical and electronic properties of the low-dimensional semiconductor structures. As an illustration, we discussed the confinement effect on the electronic properties in zinc-blend semiconductor quantum wells and presented the calculations of the optical matrix elements. The calculations were performed within the Luttinger-Kohn scheme. According to these results, one-dimensional confinement induces the splitting of the light hole- and heavy hole-bands and leads to the polarization dependence of the optical matrix elements. This is not the case, however, when the quantum well is grown in the \{100\} or \{111\} crystallographic directions. In addition, we discussed the OA dependence on the quantum well width. Similarly, the results of the calculations of the interband transition probabilities were demonstrated for the CuPt ordered crystals. In this case the light hole- and heavy hole-states mixing is due to the deformations which describe the atomic ordering. The OA investigations in the ordered crystals allow one to distinguish the ordering direction.

Finally, the methods of the observation of the OA and some recent experimental results on the anisotropy of the optical spectra in the different low-dimensional semiconductor structures were presented.

Luminescence of Spatially Confined Systems
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The electronic properties of atoms and of bulk solid state materials have been described very well by quantum mechanics. Atoms can be described in terms of localized eigenstates, and bulk solid state materials can be understood in terms of continuous delocalized bands. Spatially confined systems have electronic properties in between those of atoms and bulk solid state materials. Understanding and characterizing their properties is a real challenge. Optical spectroscopy and, in particular, luminescence spectroscopy is a very versatile tool in characterizing the electronic structure of such systems as both the spectral position of the emission (and absorption) bands and the structure of the bands strongly depend on size and nature of the spatially confined systems.

This contribution began with a general overview of theories describing the optical properties of spatially confined systems. Differences with the theories describing the atomic and bulk solid state limits were pointed out. Furthermore experiments on such materials, showing their unique behavior, determined by e.g. their size, were reviewed. Finally, possible applications were discussed.
Percolation and Localization in Semiconductor Solid Solutions  
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The following subjects were treated:

3. Valence band tail states and exciton absorption spectra.
5. Long time kinetics of exciton luminescence: the role of the conduction band tail states in exciton formation.
6. Compositional disorder in low-dimensional solid solutions: new aspects and problems to be solved.

Spontaneous Emission within a Photonic Atom: Radiate Decay Rates and Spectroscopy of Levitated Microspheres  
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Emission within a dielectric microsphere occurs preferentially into long lived modes (Q~108) which can be described by analogy with the states of an atom (photonic atom). Geometrically, the photon is confined by total internal reflection while circumnavigating near the sphere's perimeter. The microsphere provides an ideal structure for studying the emission rate and emission spectrum from a high Q meso-optic system. We presented recent fluorescence microscopy, spectroscopy, and time resolved results from experiments carried out on individual microdroplets isolated within an Electrodynamic Levitator-Trap (Paul type) and covered with surfactant flours or quantum-dots. These results show that one can alter spontaneous emission rates by changing the size of the microsphere and the orientation of emission moments near its surface. The radiation decay rate may be understood by the effect which size has on zero point fluctuations in the photonic modes of the sphere (CQED effect). This interpretation results in the determination of both the orientation and homogeneous linewidth of molecular flours at room temperature. By manipulating the alignment of the molecular structure on a sphere, one can control the gain spectrum for lasing. The fundamental properties learned through the study of droplets can be carried over to solid amorphous spheres (quartz, polystyrene). By the additional control of the density of photon states through size change one can envision the design of spherical microcavity devices (e.g. lasers, LED's, etc.) which are stimulated electrically utilizing quantum dots with radiation coupled out through an evanescently coupled optical fiber.
Coherence Effects in Quantum Optics – From Lasing without Inversion to Ultraslow Light
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Historically, coherence effects such as the Hanle effect, quantum beats, and coherent population trapping served to define the physics of the field. More recently, applications of phase coherent media have yielded electromagnetically induced transparency (EIT), lasers operating without inversion, and ultraslow light having group velocity of a few meters per second. In the near future, we will most likely have new physics and devices such as:
- Ultra high precision magnetometers and gyros using phase coherent media
- New kind of spectroscopy
- Short wavelength lasers without inversion
- New sources of IR radiation as well as new IR detectors
- Ultrashort pulses via EIT based Raman lasers
- Nonlinear optics (phase conjugate optics) beyond EIT
- Enhanced acoustic-optic effect via ultraslow light

Fiber Lasers
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Fiber lasers have excellent performance characteristics and wide applications because they possess modes confined in the coordinates transverse to the direction of propagation. This confinement essentially cancels out the effects of diffraction. With the diffraction effects canceled, long interaction lengths are possible, many meters in length if need be. Because of the long interaction lengths, even active atoms with small emission cross sections, such as Er, can provide high gains. Confined modes of fiber lasers produce other highly attractive features including very high efficiency, good transverse mode quality, compatibility with laser diode pumping, alignment insensitivity, and laser configuration flexibility. However, there are some limitations associated with fiber lasers, particularly for high energy per pulse lasers. The basic physics of fiber lasers that allow them to have these attractive features were discussed with emphasis on both advantages and limitations. Practical information on particular fiber lasers and their fabrication was presented along with examples of laser performance, both continuous and pulsed.
Structures and Models of Glasses  
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This presentation focused mostly on the study of glasses and more particularly inorganic glasses. The classical explanation for the formation of an inorganic glass is that when a liquid is cooled, its fluidity (reciprocal of viscosity) decreases and at a certain temperature below the freezing point becomes nearly zero. The relation between crystal, liquid and glass can be explained by means of a volume-temperature diagram.

The final product has all the characteristics of a solid without crystallization; we can speak of a glass as a non-crystalline solid exhibiting the phenomenon of glass transition which lacks the characteristic of sufficient internal stability. Sometimes we can also speak of a glass in term of an amorphous material. In a general way, in order to obtain a non-crystalline solid, it is necessary to block-in the structural disorder of a liquid phase. Like liquids, glasses thus possess a structure lacking long-range order. They present only short-range order which corresponds to the mutual arrangement of nearest neighbors to a given atom and varies according to the atomic site considered.

The main objectives of this lecture were to define, to characterize and to give structural models of glasses. The following subjects were treated:
1. Definition of a glass  
2. The vitreous transition  
3. Conditions for vitrification: structural theories  
4. The structural models of glasses  
5. Recent developments of glasses

Concentration and Temperature Dependence of Basic Luminescence Processes  
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The first part of the lecture reviewed basic luminescence processes in rare-earth-ion and/or transition-metal-ion doped crystals (laser materials), which are excited optically. The down-conversion luminescence and upconversion luminescence, the three main upconversion processes – Excited State Absorption, Energy Transfer Upconversion, and Photon Avalanche – were discussed with examples including different concentrations of Er ion doped crystals at different temperatures. The second part of the lecture focused on the temperature dependence of luminescence processes in rare-earth-ion doped laser crystals.
Free Electron Laser: Operation Principles
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The possibility of obtaining radiation from free electrons has been exploited for a long time with a number of common free electron devices like Klystrons, Magnetrons, Travelling Wave Tubes, and Gyrotrons all operating in the RF wavelength region. But it is with the development of the Free Electron Laser that these kinds of devices became of interest for shorter wavelengths. FELs are presently considered the natural evolution of Synchrotron Radiation Sources, especially in the spectral region presently not covered by conventional lasers, i.e. the UV and X-ray region on one side and the IR-FIR and mm-wave region (THz) on the other side. This brief tutorial attempted to give a simple description of the physical process underlying the FEL operation, starting from the emission of an accelerated particle, to derive the properties of Synchrotron Radiation. The improvements introduced by Undulator emission were described together with a derivation of the emission wavelength and of the spectral characteristic, compared to that of conventional circular synchrotron emission. The possibility of obtaining gain from the interaction of the copropagating E. M. wave and oscillating electrons was described, defining the main parameters for the FEL operation and the emitted radiation characteristics. Finally a typical FEL experimental layout was described and a certain number of applications were proposed.

Solitons in Optical Fiber Telecommunications
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In optical fibers, the soliton is a completely nondispersive pulse made possible by the small, but significant $\chi$, in silica glass. As such solitons enable one of the most technologically important applications of nonlinear optics, viz., high bit rate, ultra long distance data transmission. We introduced the elegant, fundamental physics of solitons, followed by a sketch of the corresponding transmission technology.
Interdisciplinary Lecture: Is Quantum Mechanics a Complete Theory?
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Impressive accumulation of experimental tests confirms the validity of Quantum Mechanics and extends it to a wide domain of phenomena. On the other hand, the discussion on the foundation of quantum concepts and the debate on the problem of interpretation are not over. The question whether Quantum Mechanics is a complete theory is still open. Some controversial issues were reviewed, in particular the so-called EPR paradox and the Bell’s inequalities.

Digging for the Skull of the Cyclops
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A hypothesis was presented for the origin of the Greek myth of the Cyclops, relating it to the existence of dwarf elephants on various Mediterranean islands in the time 70 to 30 thousand years ago and to the so-called dark centuries in Greek history around 1200 to 900 BC.
LONG SEMINARS

Confined Structures Based on Point Defects in LiF Films:
Optical Properties and Applications
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Research activities concerned with colour centers in alkali halide films started recently. The use of versatile, well-assessed, and low-cost fabrication techniques consisting in physical vapor deposition of LiF films combined with an electron-beam direct writing lithographic process allows the realization of optically confined active structures, like broad-band emitters, channel waveguides and optical microcavities operating in the visible. Promising results have been obtained in the generation, amplification and waveguiding of visible light in LiF films, where the efficient formation of stable laser active colour centers by low energy electron beam irradiation induces also a local increase of the refractive index. A brief overview of the investigated optical properties was provided together with a short discussion about the perspectives of applications in miniaturized light sources.

Properties of Self-Organized Semiconductor Islands Embedded in a Semiconducting Matrix
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The seminar gave an introduction to the topic of heterostructures containing semiconducting quantum dots in a semiconducting matrix, which allow the fabrication of opto-electronic devices like laser diodes. These structures can be fabricated by employing a specific epitaxial growth mode, the Stranskii-Krastanow-growth mode, that leads to island formation in a strained layer. Examples are InAs on GaAs for the near IR range and CdSe on ZnSe for the green visible range. The advantage of the technique is the high density of dots or islands within one layer and the high structural quality that can be achieved. The density of islands can even further be increased by a vertical stacking of several island containing sheets. The strain profile of islands in underlying layers leads to a preferential formation of islands on top of these in subsequent layers and improves the overall homogeneity of island sizes and distribution.

Aside from the device prospects, these structures offer a very detailed insight in the fundamental optical properties of low dimensional systems. Of particular interest are the lateral or vertical electronic interactions between the islands, excitonic and biexcitonic properties, their temperature dependency and further properties inherent to coupled low dimensional quantum systems.
We examined the example of CdSe/ZnSe more closely. In this system, which is in many respects comparable to the III-V system InAs/GaAs, the properties are governed by growth technical problems. Cd/Zn interdiffusion has to be taken into account, i.e. the islands contain generally less than 100% CdSe in their centers and the Cd distribution profile is approximately gaussian in growth direction. Furthermore, it seems to be impossible to suppress small quantum well fluctuations and Cd accumulations even when not attempting to form islands. These fluctuations already act as low dimensional confinement centers that capture excitons and lead to a very typical evolution of the photoluminescence time or temperature dependence.

The time resolved photoluminescence of a quantum dot ensemble or fluctuating quantum well allows the direct observation of lateral transport between single fluctuations or interacting islands. An exciton created in the vicinity of a local potential dip is captured and will quickly relax to its energy minimum. However, if a non-negligible tunneling probability exists it will be transferred to neighboring deeper fluctuations, causing a typical red shift of the luminescence peak with time. We thus directly learn about the interactions in the island ensemble.

In order to study the properties of single islands various methods can be used. Non-destructive methods are optical near field microscopy and microphotoluminescence. Recently, it was possible to achieve time resolved single dot spectroscopy. The life times extracted from single dot lines allow the identification of correlated peaks, in particular phonon replica and, in conjunction with autocorrelation, excited states, thus giving a new insight in the properties of semiconductor quantum dots embedded in a semiconducting matrix.

**NASA's DIAL/Lidar Laser Technology Development Program**

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Future NASA missions to enable global monitoring of the Earth's atmosphere will include measurements of ozone, carbon dioxide, water vapor, aerosols, and clouds. These are all important constituents affecting climate and atmospheric chemistry and dynamics. Carbon dioxide has been of particular interest in recent years as a result of its increase in the atmosphere combined with its importance to understanding the global carbon cycle and the global warming phenomena. NASA Differential Absorption Lidar (DIAL) and backscatter lidar techniques are leading candidates for active remote sensing of this and other atmospheric constituents from advanced flight platforms. DIAL and lidar systems operating from space will have the capability to completely map the Earth's atmosphere for these and other molecules, as well as for tracking atmospheric phenomena, such as winds and volcanic dust transport. This presentation gave an overview of our solid-state laser technology development program with emphasis on the Ho:Tm:YLF laser development to meet measurement requirements for important NASA remote sensing missions.
The luminescence emission energy and the luminescence lifetime can differ markedly for a single-dopant emission and a pair-state emission in a semiconductor host. This effect was discussed briefly and some experiments on doped nanocrystalline materials were presented.

Pair-state emission might be favorable for applications that require high luminosity (i.e. flat panel displays). Therefore, the simulation of adoped nanocrystal, in order to calculate the probability for pair-formation was discussed. In addition, a mathematical theory (Stein-Chen Poisson approximation) was presented that enables the calculation of these statistical properties in any nanocrystal, regardless of size or crystallographic structure.

The second part of the presentation dealt with typical experiments on chemically synthesized doped nanocrystals performed in our group. These include the influence of the synthesis, control of the nanoparticle properties and efforts to obtain efficient luminescent devices. Finally, experiments on single-dot spectroscopy for the purpose of biological labeling were presented and their implications discussed.
SHORT SEMINARS

Color Centers Emission in Optical Microcavities Based on LiF Films

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Controlling and modifying spontaneous emission in optical microcavities is interesting both from a theoretical and an applicative point of view in the realization of all-solid, low-threshold, high-directional light emitters and lasers. Spatial and temporal spontaneous emission modification of F₂ color centers in LiF films placed inside planar optical microcavities were reported. Angularly resolved photoluminescence measurements show a narrowing and an enhancement of the emission spectrum along the cavity axis; time resolved luminescence measurements in different geometrical configurations show a shortening of luminescence decay time along the microcavity axis.

Optical properties of II-VI semiconductor nanocrystals

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These days there is special interest in nanocrystals as biological labels. CdS nanocrystals surrounded by a thin silica shell are promising as stable biological labels because oxygen cannot diffuse through the silica layer. In a first attempt to make core/shell CdS/SiO₂ particles, tetraethoxysilane (TES) was added to polyvinylbutyral (PVB) capped CdS nanocrystals, made by mixing aqueous solutions of Cd(ClO₄)₂ and Na₂S in ethanol. Photoluminescence and absorption were studied as a function of time.

A shift in the absorption spectra to higher energies was observed. Also decolourising of the CdS dispersions was noted. The shift of the absorption spectra in nitrogen environment is much less. If the CdS nanocrystals are kept in the dark, no shift of the absorption spectra is observed. These results could be explained by assuming that photo-etching of the nanocrystals occurs. This is quite surprising because ethanol can act as a hole-scavenger.

An increase in the photoluminescence intensity was also observed when the CdS nanoparticles were exposed to light, whether oxygen or nitrogen was present. In the dark no increase in the photoluminescence was noted. This effect was explained by curing of the polymer PVB. No evidence was found for the existence CdS/SiO₂ core-shell particles.
Excitonic transitions in cuprous oxide
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Cuprous oxide is a naturally growing semiconductor with a direct gap of 2.17eV. It is well known for its excitonic features e.g. for its hydrogen-like np-series.

The talk concentrated on the investigation of l s---)np transitions, which are the excitonic analogon to the Lyman series of the hydrogen atom. The measurement is realized with differential absorption spectroscopy ("pump-probe-technique"). The signal is detected with a Fourier spectrometer since the transition energies are in the infrared spectral range.

These transitions, if measured time resolved under high excitation conditions, might be very suitable to answer a question which has discussed for about 30 years: does excitonic Bose-Einstein condensation exist or not?

Time resolved near field spectroscopy on CdSe/ZnSe quantum islands
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Semiconductors heterostructures containing quantum islands have been intensively studied in recent years because of their potential applications in the domain of optoelectronic devices. Due to the potential fluctuations in these structures, it is possible to get excitons localized in the 3 dimensions of space. In order to understand the behaviour of these excitons, we perform time-resolved luminescence experiments with a high spatial resolution. This allows us to observe directly a few excitons and to know how they locally behave. To that purpose we used a combination of Scanning Near field Optical Microscope (SNOM) and streak camera, with a resolution of a few hundreds of nm and of a few ps. We described the experimental setup and its performance, and showed as an example measurements performed on CdSe/ZnSe quantum islands. By comparing the dynamics of the sharp line caused by localized states, we can get information about the relaxation mechanisms within the sample.

Growth and electrical characterization of CdS/ZnSe heterostructures
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The conduction band offset of an CdS/ZnSe single quantum well is about 820 meV which causes a good localization of charge carriers in the CdS quantum well material. For the growth of the CdS/ZnSe heterostructures we use molecular beam epitaxy. The electrical characterization of the samples is done by Van der Pauw and Hall Bar measurements to determine the charge carrier mobility of the heterostructures.
For selectively doped systems a parallel conductivity model is suitable to describe the resulting total mobility. It can be shown that the doping concentration in the ZnSe barriers determines the total mobility. Thus the heavily doped structures are dominated by conduction in the doped ZnSe layers. By decreasing the doping concentration the characteristics of the two dimensional electron gas in the CdS quantum wells can be determined. With the decrease of the doping concentration the samples show also an anisotropic mobility, necessitating the use of Hall Bar geometry instead of the Van der Pauw method.

Science for the Masses?
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Science today has become so specialized that it is seemingly impossible for a layperson to understand any but the most basic and general of topics. However, the vast majority of people do not even understand these. I presented the (unoriginal) hypothesis that the fault lies with the scientist. We will explored the possible shortcomings of ourselves as instigators of public investigation and interest in our subjects. No consensus was reached as to possible solutions.

Growth and use of concentration gradient samples for the study of dynamical processes of laser resonant transitions in RE doped Y2O3 (RE= Yb³⁺, Er³⁺, Ho³⁺)
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The LHPG (Laser Heated Pedestal Growth) technique is a fast and efficient method to grow single crystals under the shape of fiber. Many different spectroscopic studies have already been performed on as-grown samples and laser experiments have been achieved with Nd doped Y2O3 fiber in 1973[1]. We are particularly interested in refractory sesquioxides (Y2O3, Lu2O3, Sc2O3, Gd2O3) doped with lanthanide ions (Er³⁺, Yb³⁺) for all solid state laser achievement.

A new and original process allows us to grow concentration gradient fiber by using the LHPG [2]. Composition of the dopant in these samples varies continuously from one end to the other end of the fiber between two chosen concentrations. These samples can be viewed as a collection of an infinity of single crystals of different composition and it is an efficient tool to study the dependence on composition of a physical properties.

In this short seminar we presented experimental results obtained by using concentration gradient fibers and concerning dynamical processes of resonant transition of three rare earth ions in Y2O3: Yb³⁺, Er³⁺ and Ho³⁺.
Simplified optical assembly for single-molecule spectroscopy

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A simplified modification of an optical assembly for single-molecule spectroscopy with the sample located at the joint focus of a parabolic mirror and a focusing lens was designed to fit a small helium cryostat with a 33 mm diameter neck [1]. A procedure of preadjustment of the position of the focusing lens for a known index of refraction of the environment (such as superfluid liquid helium, helium gas, or vacuum) was developed ensuring excitation of only a small area of a thin film of frozen solution; no adjustment of the lens is required during an experiment. Low impurity concentration in a sample solution and inhomogeneous broadening of the impurity spectra together with high selectivity of narrow laser excitation makes it possible to register pure-electronic spectral lines of single impurity molecules. A signal-to-noise ratio of 15:1 was achieved for single-molecule spectra of terrylene in the Shpol’skii matrix n-decane [2]. Time-dependent measurements of shape and spectral position of single-molecule lines makes it possible to obtain information on dynamical processes in solid matrices at low temperatures [2-4].


Quantum Cutting Phosphors

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Quantum cutting process involves the emission of two visible photons per each VUV photon absorbed. The application of quantum cutting phosphors in fluorescent lamps using a Xe discharge could increase the efficiency by reducing the losses during the conversion of the incident radiation into visible light.

Quantum cutting via photon cascade emission was first observed in Pr^{3+}-doped fluorides and the visible quantum efficiency of 140% was determined for YF_{3}:Pr^{3+} under 185 nm excitation [1].
Photon cascade emission of Pr$^{3+}$ was also observed in oxidic host lattices but the visible quantum efficiency was lower than 100% due to losses in UV and IR or non-radiative decay [2].

Another concept of quantum cutting is based on a combination of two lanthanide ions and involves one or two steps of energy transfer followed by two photon emission. The process was called downconversion [3]. Visible quantum cutting via downconversion was demonstrated in LiGdF$_4$:Eu$^{3+}$. The excitation into the high energy levels of Gd$^{3+}$ is followed by two step energy transfer from Gd$^{3+}$ to Eu$^{3+}$ and leads to the emission of two visible photons.

Although the efficiency of this process is 190%, an external quantum efficiency of only 32% was determined [4], mainly due to competitive absorption of the Xe discharge.

In order to optimise the quantum cutting efficiency, the Gd$^{3+}$ - Eu$^{3+}$ couple was applied in a number of fluoride host lattices and quantum cutting was also observed in NaGdF$_4$:Eu$^{3+}$, KGdF$_4$:Eu$^{3+}$, KGd$_2$F$_7$:Eu$^{3+}$ and RbGd$_2$F$_{10}$:Eu$^{3+}$. No quantum cutting was observed in K$_2$GdF$_6$:Eu$^{3+}$.

It was demonstrated that the quantum cutting effect depends on the crystal structure of the host lattice.

References:
[1] Sommerdijk et al., J. Lumin. 8, 341(1974); Piper et al., ibid., 344.

Splitting of X-ray diffraction and photoluminescence peaks in InGaN/GaN layers
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The presence of two, or more, X-ray diffraction (XRD) peaks from an InGaN epilayer is sometimes regarded as an indicator of phase segregation. In this report, detailed characterization of an InGaN/GaN bilayer by a combination of XRD and Rutherford backscattering spectrometry (RBS) showed that splitting of the XRD peak may occur in the absence of phase decomposition. An XRD reciprocal space map performed on the (105) plane showed that one component of the partially resolved InGaN double peak is almost aligned with that of the GaN buffer, indicating that part of the layer is pseudomorphic to the GaN template. From a consideration of the effect of strain on the c- and a- lattice constants, both the partially relaxed and the pseudomorphic components were shown to have the same indium content. The layer composition deduced from XRD measurements is confirmed by RBS. Depth-resolving RBS/channelling angular scans also shows that the region closer to the GaN/InGaN interface is nearly pseudomorphic to the GaN substrate, whereas the surface region is almost fully relaxed. Furthermore,
photoluminescence (PL) spectroscopy shows a double peak corresponding to the strained and relaxed regions of the sample. The presence of the XRD and PL double peaks is shown to be unrelated to compositional grading or phase separation.

Optical properties of InGaN alloys: an unsolved mystery
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The high efficiency of luminescence from InGaN has made possible the widespread recent developments in blue-green semiconductor optoelectronics. Localization of excitations, whether by composition fluctuations or self-formed quantum dots, appears to tilt the balance in favor of radiative recombination, despite the presence of huge densities of extended defects found in ‘device-grade’ material. What is not clear at the present stage is the relationship between the composition and structure; what exactly is responsible for optical effects in this material? Why does In incorporation markedly increase the luminescence efficiency? Despite much research regarding this material, we are still confronted by a mystery when considering the origin of luminescence from InGaN. In this lecture, some of the key optical and structural properties of InGaN alloys were presented, and some of the remaining open questions were highlighted.

Study of Irradiation Defects in Quantum Structures of Semiconductors A3B5
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In the past few years, quantum dot (QD) structures have been gaining increasing interest due to their outstanding performance. One of the most important promises of QD nanotechnology is the increased tolerance to defects. Higher quantum efficiency becomes possible with the basic argument that localized carriers will exhibit reduced migration to non-radiative centres. Such property is advantageous for active layers in matrix materials with a higher number of structural defects. The tolerance to irradiation-induced defects is of crucial importance in atomic energy and space applications.

The study of irradiation-induced defects in quantum structures is the main goal of my PhD thesis and I presented some results we have obtained.

The quenching of photoluminescence (PL) in InAs/GaAs quantum dots (QDs) and quantum wells (QWs) by 2 MeV electron irradiation has been investigated. We demonstrated a much higher radiation hardness of the QDs. Possible mechanisms of this phenomenon were discussed in terms of the wave function localization and defect reactions.

The influence of irradiation on the time-resolved PL of QDs and QWs has also been investigated. Both rise and decay kinetics is changed due to irradiation-induced defects. The transients prior to irradiation can be fitted with single values of the rise and

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decay time. After irradiation, the PL kinetics in the QWs still can be fitted with single (reduced) rise and decay time constants. On the contrary, the PL decay in the QD samples can only be described by at least two different time constants, one being characteristic of the as-grown sample and another (shorter) one that almost does not change with irradiation dose. Thus, we believe to have observed an interaction of confined carriers with irradiation defects inside or near the QDs.

Some future work was proposed in order to clarify some ideas related to the influence of the irradiation on the quantum structures.

Numerical Studies of Semiconductor Quantum Structures
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We have simulated the effect of strain and external fields on the electronic and optical properties of low-dimensional semiconductor structures. We have studied strain-induced quantum dots (QDs), InAs QDs, and self-organized quantum wires (QWR). The strain distribution was calculated using finite element method and elastic continuum approximation, including piezoelectric coupling. Our latest project is to calculate the band structure of the studied structures, using a 8-coupled-band model, based on the k-dot-p theory. With these calculations we intend to study the conductivity and the optical gain of QWRs and QDs.

We studied the Stark effect in strain-induced quantum dots and calculated a shift of about 1 meV in the luminescence energy for the highest field for which we still received bound states in the dot (2e-4 V/nm). In addition, we obtained a rapid decrease in the luminescence intensity for the three lowest photon emission processes at higher electric field. The numerical results are in good agreement with the experiments.

Visible luminescence of silicon microstructures fabricated with femtosecond-laser irradiation
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We reported visible luminescence from SiOx formed by microstructuring silicon with femtosecond laser pulses in air. Incorporation of oxygen into the silicon lattice occurs only where the laser beam strikes the surface. Laser-structuring therefore offers the possibility to write luminescent submicron features without the use of masks. The amount of oxygen incorporated depends on the laser fluence used for microstructuring. The peak wavelength of the primary luminescence band varies between 540 and 630 nm and depends on the number of laser shots used for microstructuring. Upon annealing, the intensity of the primary luminescence band increases significantly without any change in the luminescence peak wavelength, suggesting that the luminescence is not related to recombination of quantum-confined excitons.
Photodisruption in single cells using femtosecond laser pulses  
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When femtosecond laser pulses are focused tightly into a transparent material, the intensity in the focal volume can become high enough to cause nonlinear absorption of laser energy. The absorption, in turn, can lead to permanent structural or chemical changes. Such changes can be used to micromachine bulk transparent material such as single cells. Using a 1.4 numerical aperture microscope objective, we can photodisrupt subcellular structures inside single cells, therefore directly addressing biological questions involving such structures.

Strain and indium compositions fluctuations in InGaN/GaN wurtzite epitaxial  
films studied by Raman Spectroscopy  
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We reported on first-order Raman backscattering measurements on In$_{x}$Ga$_{1-x}$N wurtzite epitaxial layers grown by metalorganic chemical vapor deposition on the top of GaN/Sapphire substrates. The structural parameters were determined combining Rutherford backscattering spectrometry and high-resolution x-ray diffraction measurements. Considering the dependence of the band gap energy on the indium composition, x, determined in previous work [1], Raman measurements have been performed tuning excitation energy from 2.34 eV (530 nm) to 3.0 eV (413 nm). A shift of the A$_{1}$(LO) phonon mode frequency was observed, suggesting that a composition/strain variation is present. However it is found that in some cases the assumptions of the resonance in the band gap lead to incompatible interpretations about the state of strain of the sample. Therefore other resonance processes should be considered. An attempt to interpret the asymmetrical Raman line shape of this mode is made using the “Spatial Correlation” model.

Semiconductors quantum dots (QDs) possess discrete excitonic and phonon spectra. For a spherical QD of radius \( R \), the excitonic spectrum in the effective mass approximation (EMA) is well known [1]. Quantization of dipole-active optical phonons in very small crystals has received much less attention. Usually it is assumed that, for a spherical QD, there is only one far-infrared (FIR) active mode corresponding to a uniform polarization of the sphere, called the Fröhlich mode. Its frequency does not depend on the sphere's radius. This is approximately correct if the QD radius exceeds 2-3 nm for II-VI materials like CdS and CdSe. For smaller QDs, the quantization of phonon modes is important. As it has been shown theoretically and experimentally in [2], FIR absorption spectrum of such a QD is constituted by a set of (homogeneously broadened) peaks at certain frequencies within the reststrahlen band of the bulk material. These phonon frequencies in function of the QD size can be calculated within a continuum approach similar to the EMA [3] or numerically.

The FIR transmittance spectra of composite films clearly show a multimode structure instead of a single dip (due to the Fröhlich mode) observed for larger II-VI QDs [4]. This structure has been successfully explained by calculating the frequencies of confined optical vibrations with angular momentum \( l=1 \) [2].

Electromagnetic interaction between excited QDs can be important even if they are separated, as it follows from our studies of the absorption of electromagnetic waves of the FIR and optical spectral regions by composite materials containing CdSe QDs in different concentrations. The optical absorption spectrum of the most dilute film shows several peaks corresponding to confined excitonic states. It was modelled using the modified MG formalism as described in [5]. When the QD concentration increases, this structure becomes less pronounced and the peaks shift slightly to the lower energies. The similarity of these effects, which we observed in the FIR and optical spectral regions, leads us to the conclusion that the reason for both of them is the dipole-dipole interaction in dense QD ensembles.

The electromagnetic interaction leads to some broadening of the excitonic resonances like it happens, for example, to electronic levels of an atom when the atoms form a cluster. Also, the resonant peaks are red-shifted due to this interaction. Therefore we suggest that the experimentally observed modification of the absorption spectra in dense QD composites is the result of the dipole-dipole interaction between (optically excited) QDs.

Effect of the matrix on the radiative lifetimes of rare earth doped nanoparticles embedded in matrices

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The radiative lifetimes, $\tau_R$, of the excited states of rare earth (RE) ions contained in nanocrystalline insulators are different compared to their values in crystallographically equivalent bulk crystals. Their lifetimes depend on the effective index of refraction of the media consisting of nanoparticles and the substance filling the space between them. Here the radiative rates were studied as a function of particle size and the amorphous matrices containing the nanoparticles. Effects due to the effective index of refraction and site distortions associated with the increased inhomogeneous broadening were observed.

Evidence for long-range interactions of rare earth ions doped in nanocrystals embedded in amorphous matrices with two-level systems of the matrix

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The low-temperature homogeneous broadening of the electronic transitions of Eu$^{3+}$ and Pr$^{3+}$ rare earth impurity ions in $Y_2O_3$ and $LaF_3$ nanocrystals embedded into amorphous materials (polymer and oxyfluoride glass ceramics) was studied with hole-burning and fluorescence line narrowing techniques. It was shown that the homogeneous line width is determined by the interaction of the impurity ions contained in the nanocrystals with the two-level systems (TLS) of the surrounding glass matrix. A comparison of the experiments with a calculation provides direct evidence for the long-range nature of the interaction with the TLS.

Sol-gel processed Eu$^{2+}$ doped alkaline earth aluminates $MA_1O_4:Eu^{2+}$ ($M = Ca, Sr$)

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Eu$^{2+}$ doped alkaline earth aluminates $MA_1O_4:Eu^{2+}$ ($M = Ca, Sr$) show strong luminescence at the blue/green region [1]. The luminescence is characterized by a rapid initial decay from Eu$^{2+}$ followed by very long afterglow. The afterglow enhanced by codoping with some RE$^{3+}$-ions, may be due to the thermal activation of holes from traps.
followed by the emission of Eu$^{2+}$ [2]. The detailed mechanism is not, however, understood.

The sol-gel process is an efficient technique for the preparation of phosphors due to the good mixing of starting materials and relatively low reaction temperature. The solid state preparation of MA$\text{I}_2$O$_4$ is usually carried out at around 1300 °C since impurities as M$_3$AI$_2$O$_6$ are formed at lower temperatures. With the sol-gel technique, a lower temperature (900 °C) is reported for the successful preparation of MA$\text{I}_2$O$_4$ ceramics [3].

In this work, the MA$\text{I}_2$O$_4$:Eu$^{2+}$ phosphors were prepared by the sol-gel method. Thermal analysis revealed that the sol-gel technique lowered the reaction threshold temperature by 300 °C compared to the solid state reaction. However, by-products were observed in the X-ray diffraction pattern if the final heating temperature was below 1200 °C. The metastable, presumably hexagonal phase of CaAI$_2$O$_4$:Eu$^{2+}$, not formed in the solid state reaction, was obtained by the sol-gel technique at 850 °C. The structure of this phase was studied.

The luminescence of the sol-gel products was observed at the same region as the solid state prepared materials but the band of metastable CaAI$_2$O$_4$:Eu$^{2+}$ was observed at slightly higher wavelength than the normal monoclinic phase. Thermoluminescence was also studied and compared to solid state prepared MA$\text{I}_2$O$_4$:Eu$^{2+}$. Persistent luminescence mechanisms were proposed based on the results of the presented experimental work.


Nonlinear optical properties of metal nanoparticles: hyper-Rayleigh scattering studies

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Hyper-Rayleigh scattering (HRS) experiments have been used to study colloidal metal particles in aqueous solution. For solution species, HRS reports directly on the magnitude of the first hyperpolarizability $\chi$, which describes the second-order nonlinear optical (NLO) response. Substantial NLO activity is a key requirement of emerging technological applications such as photonics and all-optical switching, and the search for NLO-active materials is intense. Previous work from our lab has demonstrated the exceptional NLO response of solutions of nanometer-sized particles of gold, silver, and copper. The origin of the response is partial one- or two-photon resonance of the incident laser light used in the experiment with the particles' strong surface plasmon absorption. Particles of metals which do not display surface plasmon absorption (for example, platinum) are NLO-inactive.

As a continuation of our work on spherical metallic particles, several other interesting particle motifs have been investigated. HRS experiments on rigid, well-defined assemblies of three gold nanoparticles display enhanced NLO behavior compared to isolated single particles. The phenomenon is attributed to breaking of centrosymmetry.
on the nanometer scale: the triangular assemblies are the smallest collection of particles for which inversion symmetry is broken. Bimetallic particles are attractive targets for study, as the introduction of another metal allows tunability of the particles' optical properties. Alloy and core-shell particles have been synthesized and investigated by HRS. As with the linear optical properties, tunability of the NLO response based on composition and particle structure are observed.

Luminescence Center Excited State Absorption in Calcium and Zinc Tungsates
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The tungstate crystals are well known scintillators. The mechanism of luminescence center formation and the luminescence center model are under discussion today. The results of time-resolved spectroscopy of luminescence center in ZnWO$_4$ and CaWO$_4$ in wide temperature regions was presented. The luminescence and induced absorption under pulsed electron beam excitation (pulse duration 10 ns, 0.26 MeV) were studied. The experimental equipment used allows one to obtain the transient absorption spectra, luminescence decay kinetics and transient absorption relaxation times.

The luminescence center in both crystals represents the tungstate-oxygen complex (WO$_4^{2-}$ and WO$_6^{6-}$ in CaWO$_4$ and ZnWO$_4$, respectively). In spite of different complex symmetry the efficiency of luminescence center formation in both crystals is high. According to a preconceived idea, the luminescence is due to self-trapped excitons on the tungstate sublattice. The precursors for exciton formation are a self-trapped hole (O$^-$ type center) and an electron temporary trapped at W site. Comparison of luminescence and induced absorption life-time temperature dependencies gives strong evidence that absorption observed in Ca and Zn tungstates go from the luminescence center excited state (exciton) to some upper state. Besides the luminescence center excited state absorption, the transient absorption from trapped holes and electrons was observed in some tungstates studied.

Multi-phonon optical transitions in quantum nanostructures based on ionic crystals
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This paper deals with the theory of optical transitions in quantum nanostructures based on ionic crystals where a process of interband transition is accompanied by the creation of the electron and hole polarons. The energy of the size quantization in the quantum nanostructure exceeds in many semiconductors the electron and hole energy shifts caused by the electron-phonon interaction. In this case, the bulk semiconductor polaron radius is larger than the size of the quantum nanostructure.
Electron and hole polaron effects in nanostructures have been considered using the Kane model and the Luttinger Hamiltonian approximation. It has been shown that the hole polaron polarization effect differs from the electron polarization effect because of the degeneration of hole bands. The binding energy of a polaron increases for decreasing nanostructure dimensionality. The comparison of polaron effects in different nanostructures has shown that the maximum polaron effect is in the quantum dot. The intensity of polaron exciton absorption in a spherical quantum dot is calculated. It has been shown that the polaron effect results in optical spectra as a series of intense phonon replicas of the exciton absorption line and in the Stokes shift of absorption and emission lines. The broadening of different phonon replicas is shown to be less than their separation [1].


Strain and composition in InGaN/GaN Layers
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We investigate strain and composition of epitaxial single layers of wurtzite In$_x$Ga$_{1-x}$N (0<x<0.25) grown by MOCVD on top of GaN/Al$_2$O$_3$ substrates. It was shown that significant inaccuracies may arise in composition assessments if strain in In$_x$Ga$_{1-x}$N/GaN heterostructures is not properly taken into account. Rutherford backscattering spectrometry (RBS) measures composition, free from the effects of strain and with depth resolution. Using X-ray diffraction (XRD) we measured both a- and c-parameters of the strained wurtzite films. By measuring both lattice parameters and solving Hooke's equation, a good estimation for composition can be obtained from XRD data. The agreement between RBS and XRD data for composition allows reliable values for perpendicular ($\varepsilon_\perp$) and parallel strain components ($\varepsilon_\parallel$) to be determined. RBS and depth resolved cathodoluminescence (CL) measurements further indicate that the indium content is not uniform over depth in some samples. This effect occurs for the most strained layers, suggesting that strain is the driving force for compositional pulling.
Raman Spectroscopy Studies in InGaN/GaN Epitaxial Layers  
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In this work we studied a set of nominally undoped epitaxial In$_{x}$Ga$_{1-x}$N wurtzite films grown on (0001) sapphire substrates. In order to separate the contribution of the strain and indium content in the phonon mode frequency, indium mole fraction was determined using a strain insensitive method, Rutherford backscattering spectroscopy (RBS). Strain was evaluated by comparing the lattice constants measured by X-ray diffraction (XRD) with the relaxed lattice parameters given by Vegard's law. Samples with comparable indium content, but under different states of strain, were used as reference. This allowed the behaviour of the different Raman shift modes for both strain and composition to be independently established.

Thermal Effects of Nd PL Spectra in Garnet Hosts  
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Nd doped crystals are systems of great interests for spectroscopic studies and laser applications. Although Nd:YAG has been one of the most popular laser materials with a wavelength of 1.06µm, the study of Nd in different hosts may uncover the underlying reasons for a better efficiency and satisfy different necessities of laser fabrications. On the other hand, due to the coupling of ions to the lattice vibrations, temperature is of great relevance and plays an important role in the enfolding of the dynamic processes.
FIRST ROUNDTABLE DISCUSSION

The first roundtable discussion opened with the comment that while it is nice to have copies of all of the transparencies and/or PowerPoint slides, the amount of paper required is too much. Scanning the lecture notes into a computer and then burning a CD-ROM was the first suggested solution. It was noted that at the previous course the same problem was discussed, the proposed solution being a course website with all of the lecture notes. There was some concern about copyright infringement if lecture notes are posted on the web (especially by G. Baldacchini and his "Flash Gordon" slide). However, "fair use" and the lack of profit may still permit such a solution.

It was noted that discussion between the graduate students and the lecturers was quite low. Lecturers were reminded to be as didactic as possible. Participants were also encouraged to mix company at meals and ask questions privately if that would be more comfortable. Of course, the Marsala room was presented as a wonderful opportunity to mingle and discuss any topics of interest. In addition to the graduate students asking questions, the lecturers requested that the short seminars be placed earlier in the course. This would allow people to know which person is interested in which areas. Discussion during a lecture was encouraged.

The length of the midday break was called to our attention. It was considered too long by some and the following alternative schedule was suggested: start at 10:00 in the morning instead of at 9:00, after the morning session have a three hour break, and then maintain the afternoon schedule. This was not universally supported. A request was made for an updated map of Erice, as several restaurants labeled on the current map either no longer exist, or are not affiliated with the Center.

Monday, June 18, 2001
SECOND ROUNDTABLE DISCUSSION

The second roundtable discussion opened at 5:40pm. Professor Mazur acted as moderator. He first gave a review of the first roundtable and clarified the difference between that discussion and this discussion. The purpose of the first was to steer this course while the purpose of the second was to direct future courses. Bearing in mind that you can never make everyone happy all the time, he opened the floor for comments, questions, and suggestions.

The first comments were requests for more exercises during lectures of 3-4 hours. It was noted that a few more lecturers did add exercises after suggestions from previous years. A potential drawback brought to light was the uncertainty in the length of time required for participants to complete the exercises. However, the majority opinion was that exercises made points very memorable and easy to learn. Someone reminded the group that this was, after all, a school and learning should be the primary goal. This lead to a discussion of the topics covered in the course. The past four years have seen more chemists than physicists attend the course. Hence there was a request that more chemical based lectures be presented. On occasion, students felt lost because the material presented was too specialized. Some people disagreed and felt that the scientific content was just fine, however they felt a problem lay in the overheads. In general, overheads needs to be more readable and fewer in number. A ground rule of 15 slides for 40 minutes of lecture was recalled. Another possible problem lay in how the lecturers utilized their time. As a solution, it was suggested that there be fewer lecturers. Each lecturer would then get more time in which they would be expected to present absolutely no new work. They would only cover didactic material.

After a brief lull, a call for comments on accommodations was made. Universal support was given to the idea that participants should never be locked in San Domenico. There was also a request that the curtains in the lecture halls be open so that participants can enjoy the artwork on the walls. Everyone also agreed that there should be more choices in restaurants. There was also a request for more information about Erice itself (i.e. church mass/worship times). The washers and dryers need to be upgraded if people are expected to interact so closely for such a long time.

Some topics of future courses suggested were: 1) Scanning Probe Microscopy, 2) Single Molecule Spectroscopy, and 3) Quantum Computing. Everyone was reminded that, while all topics were good suggestions, this is a school on Spectroscopy and any course theme must reflect this fact.

Rino closed the meeting with a wonderful quote: "It is always like that in life. There is always one day when school is over. And our school is over." The meeting ended with dinner scheduled at La Pineta.
SUMMARY OF THE COURSE

R. v. Baltz

The 18th Course of the International School of Atomic and Molecular Spectroscopy focused on systems with spatially confined structures. As all previous courses, the school was organized and directed by Prof. Di Bartolo, which consisted of approximately 60 participants from Europe, North America, several countries from the former Soviet Union, and China. The main aim of the school was to bring together students and researchers from universities, research institutes, and industry to study a promising new field beginning from the basics and advance to the frontiers. ("We are all students" - the first of the "Erice-laws").

In the first part of the course a series of lectures on an introductory level was presented: States and spectroscopy of confined systems (Di Bartolo), Photonic structures (v. Baltz), Optical near field spectroscopy (Wegner), and Creation and applications of ultrashort laser pulses (Mazur).

The second group of lectures was devoted to semiconductor microstructures: Growth and spectroscopy of quantum wires (Kapon) and of Quantum dots (Elisabeth Kurtz), Theoretical concepts (Haug), Bloch oscillations (Lyssenko), Optical anisotropy (Helena Tsitsisvili) and Percolation and localization caused by compositional disorder (Reznitzky).

The third group - small but elegant- discussed the propagation and quantum properties of light: Spontaneous emission within a photonic atom (Arnold), Coherence effects in quantum optics (Scully), and Solitons in optical fiber telecommunications (Mollenauer).

The last and largest group of lectures dealt with lasers, luminescence, and materials: Future of laser technology for spectroscopy (Baldacchini), Confined structures based on point defects in LiF (Rosa Maria Montereali), Current laser research at NASA for remote sensing (J. Barnes) and of Fiber lasers (N. Barnes), Structure and models of glasses (Boulon), Luminescence of rare earth doped glasses (Xuesheng Chen), Luminescence of confined systems (Ronda), Synthesis and simulation of rare earth doped quantum dots (Suyver), and a Survey on the principle and realization of the free electron laser (Giovenale).

Many of the young participants also contributed by short seminars (10 min) and posters about their scientific work, which strongly stimulated the interaction between the attendees.

In addition to the main subject of the course we had three interdisciplinary contributions on very different subjects: About the interpretation and reformation of quantum mechanics towards a deterministic description of nature (Costa), Digging for the skull of the cyclops – the fight of the cunning Ulysses and a possible origin of the mythos of the one-eyed giants (Klingshim), and a fascinating slide-show about Southeast Asia (Mazur).

During the 14 days of the course we had the privilege of staying in a great place which – besides science – offered plenty of cultural and historical inspirations. Helena, our beautiful touristic guide, informed us about the glorious times of Selinunte and Segesta and did not fail to tell us also about the cult of Venus and the creation of “fertile energy” which was practiced in ancient times in Erice.
Four well-trained sportsmen (including two Profs.) biked up the Erice mountain (Δh=750m) in a fantastic time (70 min. pure biking time), whereas others preferred intensive dancing after dinner, the lazy ones refused any type of sports. The Marsala cellar was always the best place to meet after strenuous scientific sessions or excursions.

The writer of these lines found only but one example to complain about: The disappearance of Feynman's formulation of local conservation laws in terms of Erice-cats which – for so many years – were permanent guests close to the entrance of the Feynman Hall*. We also experienced many examples of other Erice-laws such as “there is only but one authorized source of information”, “a man has to do what a man has to do” (see bike tour), or “there is nothing so practical as a good theory” (now globally valid).

Not to forget the staff of the Majorana Center and, in particular, Mathew Vannette and his wife, Jennifer, Xuesheng Chen, and John and Daniel Di Bartolo, and Claudia Rincon for their assistance during the course and the preparation of the Proceedings which still has to be mastered. Many thanks to all of you.

A special thanks we owe to Rino Di Bartolo, who with his fatherly care, included us in his family and showed and shared with us his beloved country.

*For a reference see the article of this author in the Proceeding of the previous Erice School on Energy Transfer (1999).
SCHEDULE OF THE COURSE

**Friday, June 15, 2001**

Morning and Afternoon: Arrival and Registration at the Secretariat of the School

8:00 PM Meeting with the Director of the School

**Saturday, June 16, 2001**

9:00 – 9:50 AM Di Bartolo – *Opening Remarks*

10:30 Tour of Erice

4:40 – 5:20 PM Di Bartolo I – *The Role of Structure Confinement in the Spectroscopy of Physical Systems*

5:40 – 6:30 Di Bartolo II – *The Role of Structure Confinement in the Spectroscopy of Physical Systems*

**Sunday, June 17, 2001**

9:00 – 9:50 AM Di Bartolo III – *The Role of Structure Confinement in the Spectroscopy of Physical Systems*

10:00 – 10:50 Di Bartolo IV – *The Role of Structure Confinement in the Spectroscopy of Physical Systems*

11:10 – noon Di Bartolo V – *The Role of Structure Confinement in the Spectroscopy of Physical Systems*

4:40 – 5:20 PM von Baltz I – *Photonic Structures: Atoms, Molecules, and Crystals*

5:40 – 6:30 von Baltz II – *Photonic Structures: Atoms, Molecules, and Crystals*

6:40 – 7:30 Wegener I – *Optical Near-Field Spectroscopy*
Monday, June 18, 2001

9:00 – 9:50 AM  Wegener II – Optical Near-Field Spectroscopy
10:00 – 10:50  Wegener III – Optical Near-Field Spectroscopy
11:10 – noon  Wegener IV – Optical Near-Field Spectroscopy

5:40 – 6:30  Montereali – Confined Structures Based on Point Defects in LiF Films: Optical Properties and Applications
6:40 - 7:30  First Roundtable Discussion

Tuesday, June 19, 2001

9:00 – 9:50 AM  Mazur I – Creation, Characterization and Applications of Ultrashort Laser Pulses
10:00 – 10:50  Mazur II – Creation, Characterization and Applications of Ultrashort Laser Pulses
11:10 – noon  Mazur III – Creation, Characterization and Applications of Ultrashort Laser Pulses
12:10 – 1:00 PM  Mazur IV – Creation, Characterization and Applications of Ultrashort Laser Pulses

4:00 – 4:50  Kapon I – Growth and Spectroscopy of Quantum Wires
5:00 – 5:50  Kapon II – Growth and Spectroscopy of Quantum Wires
6:00 – 6:40  Kapon III – Growth and Spectroscopy of Quantum Wires
6:50 – 7:30  Kapon IV – Growth and Spectroscopy of Quantum Wires
Wednesday, June 20, 2001

Excursion

Thursday, June 21, 2001

9:00 – 9:50 AM  Haug I – Optical Properties of Quantum Wells, Wires and Dots
10:00 – 10:50  Haug II – Optical Properties of Quantum Wells, Wires and Dots (cont)
11:10 – noon  Baldacchini II – The Future of Laser Technology for Spectroscopy (cont)
12:10 – 1:00 PM  Baldacchini III – The Future of Laser Technology for Spectroscopy (cont)

4:40 – 5:20  Short Seminars
5:30 – 6:20  Short Seminars
6:40 – 7:30  Short Seminars

Friday, June 22, 2001

9:00 – 9:50 AM  Haug III – Optical Properties of Quantum Wells Wires and Dots (cont)
10:00 – 10:50  Haug IV – Optical Properties of Quantum Wells Wires and Dots (cont)
11:10 – noon  J. Barnes – Advances in Solid State Lasers at NASA Langley Research Center

4:40 – 5:20 PM  Lyssenko I – Bloch Oscillations in Semiconductors Superlattices
5:40 – 6:30  Lyssenko II – Bloch Oscillations in Semiconductors Superlattices (cont)
6:40 - 7:30 Lyssenko III - Bloch Oscillations in Semiconductors Superlattices (cont)

Saturday, June 23, 2001

9:00 - 9:50 AM Tsitsishvili I - Optical Anisotropy of Semiconductor Nanostructures

10:00 - 10:50 Tsitsishvili II - Optical Anisotropy of Semiconductor Nanostructures

11:10 - noon Tsitsishvili III - Optical Anisotropy of Semiconductor Nanostructures

12:10 - 1:00 PM Ronda I - Luminesence of Spatially Confined Systems

4:40 - 5:20 PM Ronda II - Luminesence of Spatially Confined Systems

5:30 - 6:20 Short Seminars

6:30 - 7:30 Short Seminars

Sunday, June 24, 2001

9:00 - 9:50 AM Kurtz – Properties of Self-Organized Semiconductor Islands Embedded in a Semiconducting Matrix

10:00 - 10:50 Reznitsky I – Percolation and Localization in Semiconductor Solid Solutions

11:00 - 11:50 Reznitsky II – Percolation and Localization in Semiconductor Solid Solutions

noon - 1:00 Reznitsky III – Percolation and Localization in Semiconductor Solid Solutions

4:40 - 5:20 PM Arnold I – Spontaneous Emission within a Photonic Atom: Radiative Decay Rates and Spectroscopy of Levitated Microspheres
5:40 – 6:30  Arnold II – Spontaneous Emission within a Photonic Atom: Radiative Decay Rates and Spectroscopy of Levitated Microspheres

6:40 – 7:30  Scully I – Coherence Effects in Quantum Optics – From Lasing without Inversion to Ultraslow Light

Monday, June 25, 2001

Excursion

Tuesday, June 26, 2001

9:00 – 9:50 AM  Scully II – Coherence Effects in Quantum Optics – From Lasing without Inversion to Ultraslow Light

10:00 – 10:50  Scully III – Coherence Effects in Quantum Optics – From Lasing without Inversion to Ultraslow Light

11:10 – noon  Scully IV – Coherence Effects in Quantum Optics – From Lasing without Inversion to Ultraslow Light

INTERDISCIPLINARY LECTURE

4:40 – 5:20 PM  Costa I – Is Quantum Mechanics a Complete Theory?

5:40 – 6:30  Costa II – Is Quantum Mechanics a Complete Theory?

6:40 – 7:30  Discussion on the subject of the interdisciplinary lecture

Wednesday, June 27, 2001

9:00 – 9:50 AM  N. Barnes I – Fiber Lasers

10:00 – 10:50  N. Barnes II – Fiber Lasers (cont)

11:10 – noon  Boulon I – Structures and Models of Glasses

12:10 – 1:00  Boulon II – Structures and Models of Glasses (cont)

4:00 – 4:50 PM  Boulon III – Structures and Models of Glasses (cont)
5:00 – 5:50  Chen I – Concentration and Temperature Dependence of Basic Luminescence Processes

6:00 – 6:50  Chen II – Concentration and Temperature Dependence of Basic Luminescence Processes (cont)

7:00 -  

Mazur V – Passage to Southeast Asia

Thursday, June 28, 2001

9:00 – 9:50 AM  Giovenale I – Free Electron Laser

10:00 – 10:50  Giovenale II – Free Electron Laser (cont)

11:10 – noon  Klingshirn – Digging for the Skull of the Cyclope

5:30 PM  Excursion

Friday, June 29, 2001

9:00 – 9:50 AM  Mollenauer I – Solitons in Optical Fiber Telecommunications

10:00 – 10:50  Mollenauer II – Solitons in Optical Fiber Telecommunications (cont)

11:10 – noon  Suyver – Synthesis and Simulation: Physical Chemistry of Doped Nanocrystals

4:40 – 5:20 PM  Summary of the Meeting

5:40 – 6:30  Round Table Discussion

6:40 – 7:30  Meeting of the Organizing Committee

Saturday, June 30, 2001

Departure