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✹ Research Report

Spectroscopy of Quantum Dots

Moungi Bawendi, David Norris, and Christopher Murray
Department of Chemistry and G. R. Harrison Spectroscopy Laboratory, MIT

With the improvement of growth techniques and micro characterization methods in the 1970's and 1980's, solid state physicists began to investigate "quantum structures." The earliest quantum structure to be fabricated and studied was the quantum well, which consists of a very thin layer (~10 nm thick) of semiconducting material, sandwiched between much thicker insulating regions (Figure 1a). When the semiconductor layer absorbs a photon and promotes an electron from the valence band into the conduction band, an electron-hole pair is formed. In the direction perpendicular to this layer, the electron and hole reside in a potential

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✹ Carl Linberger Delivers Lord Lecture

On April 9 Carl Lineberger delivered the fifth annual Richard C. Lord lecture at MIT, describing his research on "Time Resolved Dynamics in Large Cluster Ions". Dr. Lineberger is the E.U. Condon Distinguished Professor of Chemistry at the University of Colorado at Boulder, and a Fellow at the Joint Institute for Laboratory Astrophysics there. Dr. Lineberger's research is primarily experimental, using a wide variety of laser-based spectroscopic techniques to study the structure and reactivity of gas phase ions. One major thrust of this work has been to understand threshold photodetachment of negative ions, where weak, long range forces dominate the dynamics. This includes the observation of dipole-bound states of negative ions, a state where the additional electron is weakly bound in a very diffuse orbital (10's of Å) by the dipole potential of the core neutral molecule. Other goals of his work are to answer fundamental questions about structure, bonding and reactivity of ions and ionic clusters. Recent studies have been directed toward understanding the gradual evolution of chemical properties from those of an isolated molecule to a condensed phase species. Time-resolved picosecond pump-probe excitation of partially solvated ions are being used to investigate the breakup of solvent cages upon dissociation of a caged ionic chromophore.

CARL LINEBERGER

The Lord Lectureship is awarded in recognition of scientists who have made important contributions to the field of spectroscopy. This annual event is jointly sponsored by the MIT Chemistry Department and the Spectroscopy Laboratory. It was established in honor of Richard C. Lord, a distinguished physical chemist well-known for his research in infrared and Raman spectroscopy of polyatomic molecules. Professor Lord directed the Spectroscopy Laboratory from 1946 to 1976. Past Lord Lecturers have included Takashi Oka, Alexander Pines, Richard Zare and Charles Townes. ■

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✹ Personalities

DAN KLEPPNER grew up in New Rochelle, a suburb of New York City. Science excited him from an early age, but his particular interest in physics was kindled by a "great teacher" at New Rochelle High School, Arthur D. Hussey. Dan chose Williams College where a second interest—literature and poetry—was kindled. After graduating in 1953, he spent two years as an undergraduate at Cambridge University as a Fulbright Scholar. His tutor there, Kenneth Smith, was conducting research in atomic beam resonance of radioactive species and Dan became interested in atomic beams and magnetic resonance. Kenneth Smith recommended that Dan read a new monograph, Nuclear Moments, by Norman Ramsey. He also pointed out the possibility of creating an atomic clock—a subject then in its infancy—accurate enough to measure the gravitational red shift of the earth. Both of these ideas hit a resonance.

In 1955, Dan entered Harvard as a physics graduate student, and jumped at the opportunity to be a research assistant in Norman Ramsey's laboratory. Ramsey had just devised the idea of storing atoms in containers with inert walls, so that they remain polarized, providing the long interaction time needed for an accurate atomic clock. As his Ph.D. thesis project, Dan demonstrated the principle of atom storage, using Cesium in an atomic beam resonance experiment. He received the Ph.D. in 1959. The Cesium results suggested that atomic hydrogen should be an excellent candidate for atom storage. This led directly to the development of the atomic hydrogen maser by Kleppner and Ramsey in 1960, which Dan pursued first as a postdoc and then as a junior faculty member at Harvard. Success was achieved in 1960. Their very first value for the hydrogen hyperfine constant, had more than 11 significant figures (!), (The hyperfine constant is known to about 13 figures today). The direct impact of this measurement on fundamental theory was small—the theory is still struggling to achieve seven significant figures—but the hydrogen maser research led to a large body of work on atomic interactions and fundamental constants. It also eventually led to a precise measurement of the gravitational red shift by Robert Vessot at the Smithsonian Astrophysical Observatory, and the Hydrogen maser clocks have become standard equipment in other areas of science, for instance very long base line interferometry, and studies of binary pulsars.

In 1966 Dan joined the MIT faculty, where he continued his hydrogen maser studies for a number of years. He also started an experiment on atomic scattering with Dave Pritchard, who came with him as a graduate student from Harvard. Eventually Dave took over the project, made it a great success, and went on to become a fellow physics faculty member at MIT. Working with Bill Phillips, another graduate student, he measured the proton/electron magnetic moment ratio. Bill has gone on to become a pioneer in the field of laser-cooled atoms at the National Institute of Science and Technology. At about this time, Dan worked with Robert Kolenkow to develop a rigorous mechanics course, Physics 8.012, to stimulate freshmen with serious physics interest. Their book, "An Introduction To Mechanics," is widely used.

Dan's more recent interests center on the structure and behavior of Rydberg atoms — atoms excited to states of large principal quantum number (n) near the continuum. In the 1960's, Rydberg atoms with n~100 were observed in outer space, and in the 1970s Dan recognized that the new laser techniques made laboratory studies possible. These exotic atoms are huge — an n=100 Rydberg atom is nearly one micron in diameter, with an orbital area 100 million times larger than that of an ordinary atom. They have very long lifetimes, a few milliseconds for n=30. In addition, they interact strongly with electric and magnetic fields. In one Rydberg atom experiment, among the earliest in the field of cavity quantum electrodynamics , Dan demonstrated that spontaneous emission could be suppressed in a resonator with cut-off frequency below the Rydberg atom's emission frequency. Suppressed emission translates into reduced spectral linewidth, and Dan initially conceived of this technique as a means of measuring fundamental parameters, such as the Lamb shift, with improved accuracy. However it became clear that suppressing the Lamb shift linewidth also changes its frequency, an effect later demonstrated by two of his MIT colleagues, Dan Heinzen and Michael Feld.

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✹ **THE SPECTROGRAPH**

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✹ Kleppner Delivers Killian Lecture

DANIEL KLEPPNER

On March 13 Professor Daniel Kleppner delivered MIT's 1995-96 Killian Award Lecture, "Views from a Garden of Worldly Delights". Dr. Kleppner is the Lester Wolfe Professor of Physics at MIT and Associate Director of MIT's Research Laboratory of Electronics, and he conducts research in the Spectroscopy Laboratory. "Professor Kleppner's discoveries, inventions and contributions in atomic physics place him at the forefront of a science which is one of the foundations of modern technology," said the citation from the committee which selected him as this year's Killian Lecturer.

In an abstract of his talk, Dr. Kleppner said: "Science thrives by the interchange of ideas, the sharing of knowledge and the appreciation of fresh views. It is hardly a coincidence that most scientists delight in describing their work. Curing scientists of their obsession to explain would probably put an end to science. That is my rationale for talking to you about some of the views that I have glimpsed in the course of my research. I think of these as views from a garden of worldly delights, for science-embedded in nature and overflowing with wondrous creations-is indeed such a garden. I shall describe some advances in science from a rather personal point of view, tracing themes that wend through the creation of modern science and flow into today's world of atomic physics. I will explain how contemporary research on arcana such as quantum chaos and the dynamics of the vacuum is connected to the work of Johannes Kepler by a continuous stream. I do this with some diffidence, for scientists generally prefer to dream of the future rather than contemplate the past. Glimpses into the past, however, can help us put our science, and indeed our world views, into a healthier perspective. That, at any rate, is my hope." ■

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well. When the thickness of the layer is comparable to the characteristic lengths of the electron and hole, the carrier wavefunctions are quantized in the thin direction (Figure 1b). In the other two directions the carriers are not constrained. Quantum wells, for this reason, are often referred to as two-dimensional materials. A quantum dot is the zero-dimensional analog of the quantum well. In other words, a quantum dot is a nanometer-scale semiconductor structure which confines the electron-hole pair in all three dimensions (Figure 2). For cadmium selenide (CdSe), the characteristic length scale is ~112Å. If the size of the box is comparable to this length, the electron and hole are "quantum confined" by the crystallite boundary.

Our quantum dots are nanocrystallites of CdSe which are roughly spherical in shape and range in diameter from ~15 Å to ~100 Å. The electron and hole in these spherical quantum boxes have wavefunctions that are, to a first approximation, spherically sym-

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Figure 1. (a) Quantum well structure. A thin semiconductor layer is grown between thicker insulating regions. (b) Potential well formed in the conduction and valence bands in the thin direction. The electron and hole energies are quantized due to the finite size of the semiconductor layer.

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metric and therefore "atomic-like." For this reason, these materials are also often called "artificial atoms." The allure of quantum dots, or artificial atoms, is that they can teach us about the evolution of electronic properties with size as the material ranges from molecular to bulk dimensions. In addition, quantum wells have proved to have efficiencies superior to bulk semiconductors in a number of optoelectronic applications, including lasers. An unanswered question is whether quantum dots can reasonably provide further gains in efficiency. Of course, a number of obstacles remain for this question to find an answer, one of which is the attachment of electrical leads to a nanometer size crystal.

Figure 2. Quantum dot structure. A nm-scale semiconductor crystallite is embedded inside an insulating material. The potential well in Fig. 1b now applies to all three directions.

Bulk CdSe is black with an absorption edge at ~716 nm. Quantum confinement concentrates the oscillator strength in a few discrete transitions which shift blue with decreasing size as the "atomic" wavefunctions for the electron and hole are squeezed by the boundaries of the box (Figure 3). The research which we have pursued at the Harrison Spectroscopy Laboratory aims to observe and understand the evolution of the electronic spectrum of our CdSe quantum dots. Inhomo-geneities such as distributions in size and shape which conceal transitions had generally prevented observation of an excited state spectrum for these materials. Although our fabrication methods provide extremely uniform quantum dots (< 4% rms in radius), there is still enough broadening of the spectrum that absorption features are broadened and transitions concealed. We used photoluminescence excitation (PLE) studies to reduce this effect. In PLE, we optically select a subset of the sample distribution. This is demonstrated in Figure 4 with absorption and luminescence spectra for ~56 Å diameter dots. When excited well above the first transition, emission occurs from the entire sample distribution and a broad luminescence band is observed (dashed line Fig. 4a). However, when we monitor a narrow spectral band of the full sample luminescence, we optically select a subset of the sample. In the case of Figure 4a, when we monitor the fluorescence at the arrow, we are spectrally selecting the "smallest" dots in the sample. Scanning the excitation frequency while monitoring this chosen luminescence frequency gives an excitation spectrum which mimics to a large extent the absorption spectrum from the optically selected subset of the sample. This PLE spectrum (Figure 4b) has features which are significantly narrower than in the full sample absorption spectrum and this allows us to observe and monitor a number of higher excited states.

We have obtained such PLE spectra for 53 sample, ranging is size from ~24 Å to 105 Å in diameter. The data is summarized in Figure 5. The x-axis is the energy of the first excited state. Energy is more easily and precisely measured than dot size and is also a better size dependent label in this case. Using the average diameter as determined by transmission electron microscopy (TEM) as the x-axis would introduce significant size measurement error. In addition, the mean sizes obtained would not represent the subset of the size distribution which is measured in PLE. The energy of the first transition better describes those dots which are actually optically selected. The y-axis is energy relative to the first excited state. Figure 5 tracks the evolution of ten quantum dot absorption features with size. Strong features are denoted by circles and solid lines (visual guides) and weak features are denoted by crosses and dashed lines.

There are two important observations from Figure 5 that deserve attention. First, the spacing between excited states clearly decreases with increasing size. In other words, as the box gets smaller, the wavefunctions act like those for a particle in

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Figure 3. (a) Room temperature optical absorbance spectra of CdSe nanocrystallites from ~17 Å to 150 Å in diameter in increments of ~1 atomic plane. This figure shows the blue shift and discreteness of the spectrum as the wavefunctions are squeezed by the boundary of the box (the quantum confinement effect). (b) Every fifth spectrum from Fig. 3a to better display the discrete "atomic" nature of the spectrum.

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a box and their energies spread out. Second, there are two clear avoided crossings which would not be expected from simply squeezing spherical wavefunctions. The states labeled as (g) and (e) repel each other, as do states (e) an (c). These avoided crossings are like fingerprints for testing theoretical models of the evolution of the states. These features are because the crystallites are not empty boxes, but contain atoms which have complex electronic structures. We found that a theory that takes into account the underlying atomic structure (mostly the spin-orbit interaction on the Se atoms) and uses a bulk Hamiltonian with spherical boundary conditions for the walls of the box reproduces the data, including the avoided crossings. It is remarkable that these structures which contain 100's to 1,000's of atoms can be described nearly quantitatively by a fairly simple model which treats the electron and the hole as if they were free particles in a spherical potential. The wavefunctions in this model are spherically symmetric and delocalized over the volume of the crystallites, confirming the label "artificial atom."

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Figure 4. (a) Absorption (solid line) and photoluminescence (dashed line) spectra for ~56 Å diameter quantum dots. The downward arrow marks the emission position monitored in PLE. (b) PLE scan for ~56 Å diameter dots.

Figure 5. Transition energies (relative to the first excited state) vs. the energy of the first excited state. Peak positions are extracted from PLE data as shown in Fig. 4. This plot summarizes all of our PLE data. Strong (weak) transitions are denoted by circles (crosses). The solid (dashed) lines are visual guides for the strong (weak) transitions to clarify their size evolution.

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Jacques Van Dam, Brigham and Women's Hospital

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March 12	**Peter Hess**, University of Heidelberg In situ Spectroscopy of Semiconductor Growth with Monolayer Resolution
April 2	**Jeffrey I. Steinfeld**, Chemistry Department, MIT Spectroscopy of Explosive Molecules
April 9	**Fifth Annual Richard C. Lord Lecture** **Carl Lineberger**, University of Colorado Time Resolved Dynamics in Large Cluster Ions
April 16	**Klaasjan van Druten**, Physics Department, MIT Atoms in Intense Light Fields: Beyond Fermi's Golden Rule
April 23	**Philip H. Bucksbaum**, University of Michigan Strong Field Quantum Control
April 30	**Mark G. Raizen**, The University of Texas at Austin Atomic Motion in Nonlinear Potentials
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✹ Research Report

Raman and FT-IR Spectroscopy of Heteropolymer Gels

K. Tanaka, V.B. Kartha and R.R. Dasari, C. Wang, T. English, T. Tokuhiro and T. Tanaka
George R. Harrison Spectroscopy Laboratory and Department of Physics, MIT

Polymer gels can undergo a phase transition in which they change their volume by a factor of many thousands in response to environmental changes such as temperature, solvent and electric field. The phenomenon is observed in various natural and synthetic gels. Such gels are called smart gels, and many groups are trying to develop smart gels tailor-made for a variety of applications. In spite of their technological importance, there have not been many studies to understand the basic relationship between the molecular structure and interactions of such systems and their macroscopic phase behavior.

Raman and infrared spectroscopy techniques should provide ideal methods to probe the interactions leading to the macroscopic phase behavior, since the vibrational frequencies are sensitive to both intra and intermolecular interactions. Hydrophobic, hydrogen bond and electrostatic interactions manifest themselves in frequency shifts, band widths ,band shapes, and depolarization ratios because of changes induced in transition moments, local symmetry and bond force constants. In the present work we have investigated temperature-induced volume phase transition in N-isopropyl acrylamide (NIPA) polymer gels through Raman and FT-IR spectral studies, and the results are presented here.

The gels were prepared by methods described earlier [Hirotsu, 1987]. Raman spectra were recorded on a Spex 0.6 m monochromator. The 407 nm Kr^+ laser line was used for excitation at about 100 mW power, and Raman signals were detected with a Princeton Instruments liquid nitrogen-cooled CCD detector. Infrared spectra were recorded on a Perkin Elmer 1700X FT-IR spectrometer.

The Raman and IR spectra of N-isopropylacrylamide, NIPA polymer and NIPA gel at room temperature are very similar to each other, except for the $=CH_2$, =CH, and C=C bands exhibited by the monomer, in addition to the bands present in the polymer and gel. These spectra show the *trans* conformation of the amide, and indicate that the conformation is the same for the isopropyl group in all three cases. When the temperature of the sample is varied from 20 to 50^0C, the monomer spectrum is unchanged over the entire spectral range, showing that there are no conformational changes taking place. This is expected, since the barrier for rotation of the isopropyl group can be quite high and it retains the stable room temperature conformation. On the other hand, both the linear polymer and the gel show spectral changes in this temperature range.

Figure 1 shows the Raman spectra of the C-H stretch region for the monomer, polymer and gel. The CH_3 asymmetric stretches (out-of-phase and in-phase) at 2994 and 2950 cm^{-1} remain constant up to about 36^0C, and then suddenly decrease by about 5 cm^{-1} for both the polymer and the gel.

Figure 2 shows the FT-IR spectra of all the three samples for the low and high temperature range in the 1500-1000 cm^{-1} region. Again it can be seen that while the monomer spectra remain unchanged for the two temperatures, the polymer and gel show considerable change in the isopropyl C-C stretch region [Lin-Vien, 1991] at 1170-1120 cm^{-1}.

It can be seen from the spectral changes at the transition temperature that both the polymer and gel undergo a structural change which appears to be the same for both systems. Since the changes are seen only for the isopropyl bands, it can be concluded that

Figure 1. Raman spectra of NIPA monomer (A), polymer (B), and gel (C) in the isopropyl C-H stretch region. ——— 20^0 C; ..... 45^0C.

Raman and FT-IR Spectroscopy …

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the change involved is most probably a conformational change of this group. The linear polymer most probably has a coil structure, with the isopropyl groups extending out from the coil. The coil-to-globule transition of the polymer constrains this group to be displaced from the lower temperature equilibrium position because of hydrophobic interactions, and this leads to the observed spectral changes.

Spectral studies as a function of concentration for dimethyl acrylamide gels have shown no changes as the concentration is increased from about 5% to 75%. It is therefore unlikely that the observed changes in NIPA gels are from interchain interactions. The present studies thus show that the volume phase transition of the gel is very similar to the polymer transition. Further studies with isotopically labeled compounds will unequivocally identify the groups involved in the structure changes. Model calculations with different conformations are also being carried out to understand these changes at a molecular level.

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Figure 2. FT-IR spectra of NIPA monomer (A), polymer (B), and gel (C). ——— 20^0 C; ..... 45^0C.

Personalities

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Dan is currently using Rydberg atoms to study "quantum chaos", in which the behavior of the Rydberg atom's outer electron displays phenomena on the border between quantum mechanical and classical descriptions. Dan also works at the polar opposite of chaos—high—precision measurements. With his students he is measuring the Rydberg constant using, naturally, Rydberg states of hydrogen. Dan also collaborates with Tom Greytak, another MIT Physics Department colleague, to produce high densities of spin-polarized hydrogen. They hope to demonstrate Bose-Enstein condensation in atomic hydrogen, and have recently found a method to study the optical spectrum of hydrogen to much higher resolution than previously possible.

Writing continues to be a source of enjoyment for Dan. His occasional essays for Physics Today span subjects as diverse as science policy, Count Rumford, and the pain of making mistakes. In addition, he is interested in science policy, and has been active on numerous committees of the American Physical Society and the National Academy of Science. One of his essays in Physics Today is entitled, "Confessions of a Committee Junkie".

Dan lives in Belmont with his wife Beatrice, a teacher. They work together on a summer seminar which each year takes 20 undergraduate and graduate students to Japan to meet with their counterparts. They have two sons and a daughter, and Dan has just become a grandfather. ■

✹ Spectroscopy Laboratory Publications

"An Approach to Single Molecule Detection Using Surface-Enhanced Raman Scattering (SERS)", Kneipp K, Wang Y, Kneipp H, Dasari RR, Feld MS. Proceeding of International Workshop on Single Molecular Detection: Basis and Applications in Life Sciences, BiosQuant GmbH, Berlin-Adlershof, Germany, October 4-6, in press (1995).

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