THE SPECTROGRAPH

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George R. Harrison Spectroscopy Laboratory

Massachusetts Institute of Technology

Dasari Honored at Indian Institute of Technology, Kanpur

Ramachandra Dasari

On February 28, 2005 the Center for Laser Technology/Laser Technology Program at the Indian Institute of Technology (IIT), Kanpur created a distinguished lecture series in honor of Dr. Ramachandra Rao Dasari, Associate Director of the Spectroscopy Laboratory. The inaugural function was a day long program of talks from professors and senior scientists who were former IIT, Kanpur graduates and a poster session from the current students of IIT, Kanpur. Dr. Dasari spoke on “Spectroscopy for diagnosis of disease”. In each subsequent year

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Also in this issue

Spectroscopy Laboratory to Get New Physical Plant

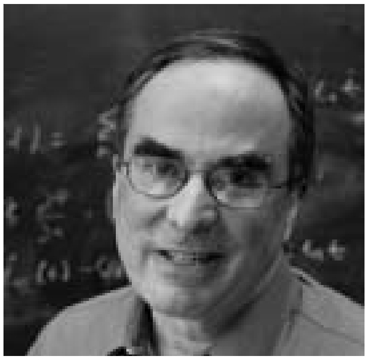
The Spectroscopy Laboratory will undergo a complete renovation over the next 18 months and aquire a new physical plant. The new laboratories and offices will be located in Bldg. 6, adjacent to the present location in Bldg. 6A. The architectural design of the 10,000 sq. ft. of new research space, with a total of 14 laboratories and two small service facilities for chemical preparation and cell culture, reflects the research directions set forth in the Laboratory’s 20 year research plan. The Laboratory last underwent renovation in 1980.

The current Spectroscopy Laboratory building, Bldg. 6A, was designed in 1928 by Karl T. Compton, MIT’s ninth president, and George R. Harrison. As a centerpiece of the newly reconstituted Physics Department, President Compton wanted to create a modern laboratory dedicated to spectroscopy and devoted much personal attention to the construction details. In an article describing the construction of the Spectroscopy Laboratory, Compton wrote that the building “expresses a definite conviction that the science of spectroscopy is destined in the future to play a role of increasing importance in bringing about an understanding of the electrical structure of atoms and molecules and the interatomic and molecular forces which underlie the interpretation of physical and chemical phenomena.” [1,2] The 13 room Spectroscopy Laboratory was completed in 1930, a full year ahead of the Eastman Laboratories (Bldg. 6). A key feature was a 10 m by 10 m walk-in spectrograph for long optical path length studies of weak spectral lines with high resolution. (Figs. 1-3). This building is believed to be the first building constructed exclusively for the purposes of spectroscopy.

Building 6A has housed the Spectroscopy Laboratory for 75 years. It is a remarkable

Rennovations, continues on page 6

Robert Field to give Lord Lecture on April 26



Robert Field

Robert Field, a Professor of Chemistry at MIT, has been selected as the 2005 Lord Lecturer. He will speak on April 26 on the topic “Just Small Enough”.

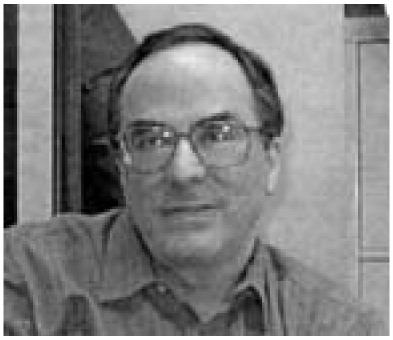
Dr. Field, a physical chemist, has a long and distinguished research career in studies of perturbations in the spectra of diatomic molecules, and he has pioneered in the development of numerous optical methods to probe the structure and dynamics of molecules. He is also the inventor of the technique of stimulated emission pumping, a widely used method for studying the electronic structure of molecules.

Now in its fourteenth year, the Lord Lecture commemorates the achievements of Richard C. Lord, a pioneer in infra-red and biochemical spectroscopy and director of the Spectroscopy Laboratory for 30 years. Each year’s lecturer is selected by a committee of chemists, physicists and engineers at MIT who are active in various fields of spectroscopy. Past recipients include Watt Webb, Charles Townes, Carl Lineberger, Steven Chu, and Britton Chance.

A biographic sketch of Dr. Field can be found on page 2.

Personality

Robert Field



Robert Field

Robert Field grew up in Chicago. His earliest memories involve 78 rpm records (Leadbelly) and food (bacon), not chemistry, although his father Edmund was a physical chemist (totally innocent of quantum mechanics) at Standard Oil of Indiana. He remembers dissolving in tears when his mother Kay excitedly told him that he had "broken a record", unaware of the distinction between black vinyl and accomplishments.

THE SPECTROGRAPH

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The Spectroscopy Laboratory houses two laser research resource facilities. The MIT Laser Research Facility, supported by the National Science Foundation, provides shared facilities for core researchers to carry out basic laser research in the physical sciences. The MIT Laser Biomedical Research Center, a National Institutes of Health Biomedical Research Technology Center, is a resource center for laser biomedical studies. The LBRC supports core and collaborative research in technological research and development. In addition, it provides advanced laser instrumentation, along with technical and scientific support, free of charge to university, industrial, and medical researchers for publishable research projects. Call or write for further information or to receive our mailings.
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The mid 1950's was too late to obtain the ingredients for explosives from chemistry sets, so Robin (that was his pre-Bob nickname) started a notebook describing the colors he was able to generate with his safety-minded chemicals. A budding spectroscopist! In sixth grade he made what was probably the most important transition of his life, from the Chicago Public Schools to the University of Chicago Laboratory School, where the teachers seemed to be genuinely interested in the ideas of their students. There were many great teachers at the Lab school, and of course one of them was a chemist.

Bob majored in chemistry at Amherst College, working with an inorganic chemist, Cooper Langford, who "set the hook" by doing a group theoretical derivation of a selection rule on a scrap of paper. But Bob's interest in spectroscopy was born during two summers in the inert gas fluoride group at Argonne National Laboratory, where he recorded and attempted to interpret proton and fluorine NMR spectra of SbF_5 in anhydrous HF. His Amherst Thesis was on UV and NMR spectra of Pd complexes of aniline (which he remembers distilling outside of a hood).

Bob's application to graduate school specified theoretical inorganic chemistry as his area of interest. By accident (or extraordinary good luck!) Bob ended up in Bill Klemperer's research group at Harvard University, where his interest in spectroscopy grew into a passion for small molecules, multiple resonance spectroscopy, spectroscopic perturbations, and other arcanae. His thesis project, measurement of the electric dipole moment of the CO $A^1\Pi$ state by radiofrequency-optical (pre-laser!) double resonance (RFODR), was a failure. However, his fits of all extant spectroscopic data on the excited valence states of CO, intended as a basis for predicting the Δ-doublet transition frequencies he needed for his RFODR-Stark effect experiment, became the foundation of his scientific career. This takes the familiar story of a warm-up project becoming *the* Thesis project to a new level! His book, "Perturbations in the Spectra of Diatomic Molecules" and its sequel, "The Spectra and Dynamics of Diatomic Molecules", his interest in semi-empirical models built on matrix element quantum number scaling rules (both atomic-ion-in-molecule ligand field theory and multichannel quantum defect theory for e^-<–>MX^+ scattering), and his optimism that an elegant simplicity is hidden behind the apparent complexity of a spectrum, all grew naturally out of his pre-thesis warm-up project.

Bob's first project as a postdoc at the University of California at Santa Barbara with Herb Broida and David Harris was to record microwave optical double resonance spectra of BaO. This laser counterpart of his RFODR Thesis project worked within two weeks of his arrival at UCSB, providing wonderful compensation for 6 years of failure at Harvard. BaO and all of the alkaline earth monoxide and monohalide molecules quickly joined CO as Bob's favorite molecules because they gave him a new use for his beloved spectroscopic perturbations. The chemiluminescent Ba + N_2O reaction had a >30% quantum yield for BaO in the metastable $a^3\Pi$ electronic state. The perturbed levels act as a funnel through which population must flow *en route* to levels of the $A^1\Sigma^+$ state that can fluoresce to the $X^1\Sigma^+$ ground state. The (short-lived) excitement over the possibility of building an electronic transition chemical laser based on the BaO $a^3\Pi \sim A^1\Sigma^+$ perturbations contributed to Bob's invitation to join the MIT faculty as Assistant Professor of Chemistry in 1974.

Bob's favorite description of his MIT research is "spectroscopy beyond molecular constants." Spectra encode dynamics and the greatest challenge is not to fill journals with archival data but to uncover a larger picture in which coupling mechanisms between intramolecular modes are characterized. How does energy flow from one bond to another, from an electron to nuclear vibrations and rotations? Simple molecules at high levels of internal excitation exhibit complex dynamics. The usual small-displacements-from-equilibrium models fail utterly because modes usually considered as separable are recoupled in unforeseen ways and fundamental concepts like chemical bonds and oxidation states can no longer be relied upon. When is a spectrum intrinsically unassignable (ergodic) and when is it merely inconvenient to use traditional assignment and spectroscopic modeling methods? Are there non-traditional methods to trick molecules into revealing their dynamical secrets?

Two molecules, acetylene and CaF, have been the subject of extraordinary experimental effort in Bob's research group. More than 100 person-years have been in-

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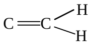
vested in the spectrum of acetylene. Bob did his first experiments on CaF at UCSB in 1973! These molecules are *just simple enough* to contain hints of fundamental dynamical processes in their spectra.

Acetylene (and its molecularly impoverished cousins HCN and HCP) can undergo bond-breaking isomerization to  vinylidene and, because vinylidene is the transition state for $H_a C_1 C_2 H_b$<–>$H_b C_1 C_2 H_a$ identical atom permutations, permutation splittings in the symmetric double minimum acetylene potential energy surface selectively label the highly excited vibrational levels that sample the minimum energy isomerization path. This is a novel way to recognize the isomerization-relevant features in a spectrum without attempting to make secure vibrational assignments of every observed transition. This is an unconventional, and still not achieved, spectroscopic (rather than chemical kinetic) way to characterize a "transition state".

CaF is the sodium atom of diatomic molecules. It contains one electron outside the Ca^{2+} and F^{-} closed-shell atoms. The details of the CaF Rydberg spectrum reveal the mechanisms of a light electron undergoing inelastic scattering off of two heavy nuclei, just as the Rydberg spectra of Na reveal the beyond-hydrogen mechanisms of an electron scattering off of a round but space-filling Na^{+} nucleus. Of course the $e^{-}\Sigma Ca^{2+}F^{-}$ scattering is much more complicated and the dynamics much richer than the $e^{-}\Sigma Na^{+}$ scattering. The goal, says Bob, which seems within grasp, is to obtain a *complete* dynamical picture, including dissociation and ionization processes, expressed in the form of an internuclear distance dependent quantum defect matrix, $\boldsymbol{\mu}(R)$.

Frequency-domain spectroscopists are collectors of eigenstates. Eigenstates are stationary, i.e. a molecule in an eigenstate does not move, but eigenstates encode dynamics. The facilitators of the most interesting intramolecular dynamics are off-diagonal matrix elements of the molecular Hamiltonian between Born-Oppenheimer basis states. Bob was amused to discover that, from his very first experiments in the Klemperer group, he was studying dynamics. Throughout his career, he has attempted to discover new ways that dynamics are encoded in spectra and new ways to record spectra that are uniquely selective to specific classes of intramolecular dynamics. The game is still afoot.

Dasari, continued from page 1

a day long program will be held in which a distinguished scientist is honored. February 28 is National Science Day in India, as well as C.V. Raman's birthday.

Dr. Dasari was born and raised in India. He was educated at Benaras Hindu University and Alighar Muslim University and is one of the first faculty members to join IIT, Kanpur at its inception. His connection to MIT dates back to 1966 when he spent two years working in the research group headed by Professor Ali Javan. His experience in lasers gained at that time helped him to set up one of the largest laser laboratories at IIT, Kanpur where lasers and laser instrumentation was fabricated and graduate students trained. During that period, as a member of the University Grants Commission, he helped improve undergraduate science programs in degree-granting colleges. He was also instrumental in the transfer of laser technology to industry. He left IIT, Kanpur in 1978, moving to the National Research Center in Ottawa for a year and the University of British Columbia for another year, before joining the MIT Spectroscopy Laboratory in 1980.

Dr. Dasari has played a major role in the Spectroscopy Laboratory over the past 25 years, coordinating research programs and facility development associated with the NIH supported Laser Biomedical Research Center supported by NIH and the NSF supported Laser Research facility. His research activities cover a wide range of disciplines: atomic and molecular physics (cavity-QED, single atom laser, atomic and molecular collisions, laser-nuclear studies); chemical physics (vibrational-rotational relaxation, Dicke narrowing in infrared transitions, laser spectroscopy of rare-earth ions); classical spectroscopy (high resolution spectroscopy of simple molecules); surface enhanced Raman scattering (single molecule detection); and his relatively large efforts in the last ten years related to laser biomedical studies, leading to spectral diagnosis of disease using techniques including scattering, reflectance, fluorescence, and Raman spectroscopy. His most recent research relates to low coherence interferometry for detecting nanometer motions in cells and nerve bundles. His research publications in referred journals number more than 200 and cover most of the major journals in physics and chemistry.

Spectral Lines

Archimedes' Solar Weapon

by Stephen R. Wilk

Stephen Wilk

The Discovery Channel's series *Mythbusters,* on cable TV features a group of Hollywood special effects experts running tests and demonstrations to verify or disprove "urban legends" and odd reports. An episode of their second season, first broadcast in September 2004, they examined the case of Archimedes' Mirror.

Archimedes is the third century B.C.E. mathematician and engineer from Syracuse in Sicily. Only a few of his works have come down to us, but those seem remarkably ahead of their time. In *The Sand Reckoner* Archimedes outlines a system for writing very large numbers, much like our system of exponentials, and then goes on to calculate how many grains of sand it would require to fill the universe. His work *On Floating Bodies* gave us Archimedes' Principle, that the weight of a body placed in water is lessened by the weight of water displaced. From his work on the circle he obtained a value of *pi* between 3 10/71 and 3 1/7.

Archimedes was also supposed to have created exotic weapons for Hiero II, the ruler of Syracuse, for use against the Romans during the Second Punic War. Of all the weapons used – catapults, siege towers, giant cranes – the most interesting is a giant mirror or collection of mirrors said to have been used to burn enemy ships. Lucian of Samosota and Galen both mention this, as does the sixth century architect of He Hagia Sophia (usually called Saint Sophia), Anthemius of Tralles, who wrote that Archimedes employed many mirrors. Proclus, a noted mathematician, was said to have also used burning mirrors against Vitellius in 514 C.E.

Can the story be true? The Mythbusters team investigated, going so far as to build a section of an ancient boat and manufacturing a sort of giant Fresnel mirror out of indi-

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vidual mirrors measuring about a foot square, placed on a large plywood backing. A depth gauge was used to adjust the tilt of each mirror segment. The resulting giant mirror focused sunlight onto an area about 2-3 feet in diameter on the section of a ship, located perhaps 100 feet away. Although the illuminated spot got warmer, it didn't get hot enough to burn, and they concluded that, in the words of the show, "The myth was busted!" I addition to their failure, they cited the unlikely conditions – you'd need a sunny day to use the solar death ray, and an enemy willing to attack then[1,2].

But was the test fair? The use of the crude focusing mechanism meant that the beam was not optimally focused. If some optical means had been used rather than mechanical means, would they have been able to kindle a fire?

Twenty two years ago Albert C. Claus of Loyola University suggested that Archimedes could have performed his feat if he had a team of soldiers armed with mirrors, each of which could be independently pointed [3]. He even proposed a method of pointing each mirror in the array[4]. O. N. Stavroudis wrote back to note that the idea of using burning mirrors against one's enemies predates Archimedes, and can be found in Aristophanes' comedy *The Clouds* (produced in 423 B.C.E.). Furthermore, all the sources for Archimedes' feat date from centuries after his time. If there was no credible account of the event, why should we believe that it happened[5]?

As K.D. Mielenz pointed out, it appears that the experiment had been done before – George Louis leClerc, Comte Buffon , famous for his "dropping needles" statistical method for determining the value of *pi*, had performed a very similar experiment[6]. Buffon assembled 168 mirrors 8 inches by 10 inches in dimension, adjusted to produce the smallest possible image 150 feet away. The array turned out to be a formidable weapon. At a distance of 66 feet, 40 mirrors ignited a creosoted plank and at 150 feet, 128 mirrors ignited a pine plank instantly. In another experiment, 45 mirrors melted six pounds of tin at 20 feet. Furthermore, in 1973 a Greek scientist, Ionannis Sakkas, got a team of 60 soldiers together, gave each a mirror, and had them direct the sun's rays at a boat 160 feet away. They succeeded in setting the boat afire. The incident was reported around the world, in the London Times, Time magazine, and the New Scientist[7].

D.L. Simms wrote critiques of the issue, concluding that it was not reasonable to believe that this was possible. Simms notes the work of Sakkas and Buffon, yet concludes rather surprisingly that it is irrelevant, since it represents a "static situation", and that it is difficult to adjust the focus properly under "battlefield" conditions[8]. Yet the focusing method suggested by Claus in that first Applied Optics report would give precisely the means to maintain a focus on a moving object – it's not necessary to know what focal length is required, as long as each independent element is capable of tracking the target.

Archimedes' Mirror

Archimedes' mirror has shown up in textbooks on adaptive optics as the earliest recorded case of a "rubber mirror"[9]. Michael Lahanas' website [7]cites a 2002 experiment in Germany in which 500 volunteers used 45 cm x 45 cm mirrors to ignite the sail of a ship at a distance of 50 meters. The test conditions seem very similar to those used on the *Mythbusters* show, except perhaps in using the sail rather than the hull of the ship as a target. The only other difference seems to be the use of volunteers for active focusing.

It's not clear whether or not Archimedes (or Proclus, or anyone else) ever actually used solar mirrors as a weapon of war, but it seems very likely that they *could* have been used succesfully [10]

References

1.Mythbusters Pictures Season 2, Episode 16, first broadcast September 29, 2004: http://dsc.discovery.com/fansites/mythbusters/photogalleries/season_02/season_02.html

2. More Mythbusters pictures: http://dsc.discovery.com/fansites/mythbusters/photogalleries/season_02/season_02_02.html

3. Claus, A. C. "On Archimedes' Burning Glass", Applied Optics 12 (10), A14 (1973).

4. In 1942 L.L. Young of the National Bureau of Standards and W.M. Potter of General Electric developed the method of aiming a heliograph described by Claus in his article. It was incorporated into signalling kits intended for downed pilots manufactured by GE. It is described in two articles by R. S. Hunter: J. Optical Society of America 35, 805 (1945); "Heliographic Signaling Mirrors", by R.S. Hunter in J. Optical Society of America 36 (2), 110-115 (1946). The same method of signalling was used in Hal Clement's science fiction novel "Cycle of Fire", (1957). Arthur C. Clarke described a similar situation in his short story, "A Slight Case of Sunstroke", which appeared in Galaxy Science Fiction in September, 1958. It subsequently appeared in his collection of short stories, "Tales of Ten Worlds" (1962). A Webpage performing the calculations is: http://zebu.uoregon.edu/~dmason/probs/therm/dyn/sunstrok/sunstrok.html

5. Stavroulis, O.N. "Comments on: On Archimedes' Burning Glass", Applied Optics 12 (10), A15 (1973).

6. Mielenz, K.D. "That Burning Glass", Applied Optics 13 (5) A14 (1974).

7. An excellent site with many references is http://www.mlahanas.de/Greeks/Mirrors.htm

8. Simms, D.L. "More on That Burning Glass of Archimedes", Applied Optics 13 (5) A16, (1974); at much greater length in D.L. Simms, "Archimedes and the Burning Mirrors" of Syracuse, Technology and Culture 18 (1), (1977).

9. Hardy, J. W. "Active Optics: A New Technology for the Control of Light", in "Archimedes' Mirror as Adaptive Optics", Proc. IEEE 66, 651 (1978); J .E. Pearson in Conference on Laser and Electro-Optical Systems, San Diego CA (1980); both cited in Principles of Adaptive Optics (2nd Edition) R. K. Tyson, Academic Press, 3.

10. Archimedes' Mirror is described in J. D. Walker's, "The Flying Circus of Physics "(Wiley, 1975) , entry 3.76 on p. 65. Curiously, in this heavily-footnoted book, no references to Archimedes Mirror are given. In the revised edition, "With Answers", published in 1977, some explanations are given on p. 247, but without citations.

Research Report

Interaction between Electronic and Rotational Motions in Calcium Monofluoride

Jeffrey J. Kay, Serhan N. Altunta, Stephen L. Coy, and Robert W. Field
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Molecular Rydberg states are interesting from a fundamental point of view because they allow us to understand, in a ball and spring sense, how energy and angular momentum are exchanged between light electrons and heavy nuclei. By their very nature, Rydberg series sample electronic motion over a wide range of timescales: along a Rydberg series, the frequency of electron orbital motion varies from just a few femtoseconds (timescale of molecular vibration) at low principal quantum number to tens of picoseconds (timescale of molecular rotation) near the series limit. By understanding how Rydberg series evolve from low to high energy, and how different series interact with each other, we can understand how electronic, vibrational, and rotational motions influence each other on a variety of timescales and identify the mechanisms that facilitate intramolecular energy transfer.

The interaction between electronic and rotational motion, with which we are concerned here, is not a new topic. It has been studied since the early 20th century and began with the work of Friedrich Hund, who categorized molecular electronic states according to various angular momentum coupling schemes, which are now known (and dreaded) as "Hund's cases". Hund's cases have been used for nearly 80 years to classify electronic states and to define basis sets by which complicated dynamical problems may be described. Two of Hund's cases are especially relevant to the description of molecular Rydberg states and offer pictorial views of two limiting cases of rotation-electron coupling. Energetically low-lying Rydberg states typically conform to Hund's case (b), in which the angular momentum of the Rydberg electron precesses about the internuclear axis of the molecule under the influence of the cylindrically-symmetric electric field of the nuclei. Consequently, neither the orbital angular momentum l of the Rydberg electron nor the rotational angular momentum N^+ of the nuclei ("ion core") are strictly conserved; only the total angular momentum of the molecule N is a constant of the motion. The two particles interact strongly through the electric field of the ion core and the molecule "rotates as a whole". When the electrostatic interactions between the Rydberg electron and the ion core become sufficiently weak, Rydberg states conform to Hund's case (d), in which both particles move more or less independently of one another. As a result, these Rydberg states can be described in terms of the quantum numbers of the separate particles; under these circumstances both the orbital angular momentum l and rotational angular momentum N^+ remain well-defined.

Despite almost eight decades of research on the topic, a simple picture of how electronic and rotational motion interact on *all* time scales remains elusive. In particular, it is not clear how electronic and ion-core rotational motions influence each other when the interactions involved are *strong*. In the low-lying Rydberg states of calcium monofluoride, a molecule with a long history in our research group, the electrostatic interactions between the electron and ion are so strong that these states are very well described in terms of Hund's case (b) coupling. Since the low energy Rydberg states cannot be described in terms of the separate angular momenta of the separate particles, it is not obvious how these Rydberg series will evolve with increasing principal quantum number, especially when the bound Rydberg states merge into their respective ionization continua. One might imagine that, as the principal quantum number increases and the time scale of electronic motion becomes longer and the average dis-

Interaction, *continues on page 6*

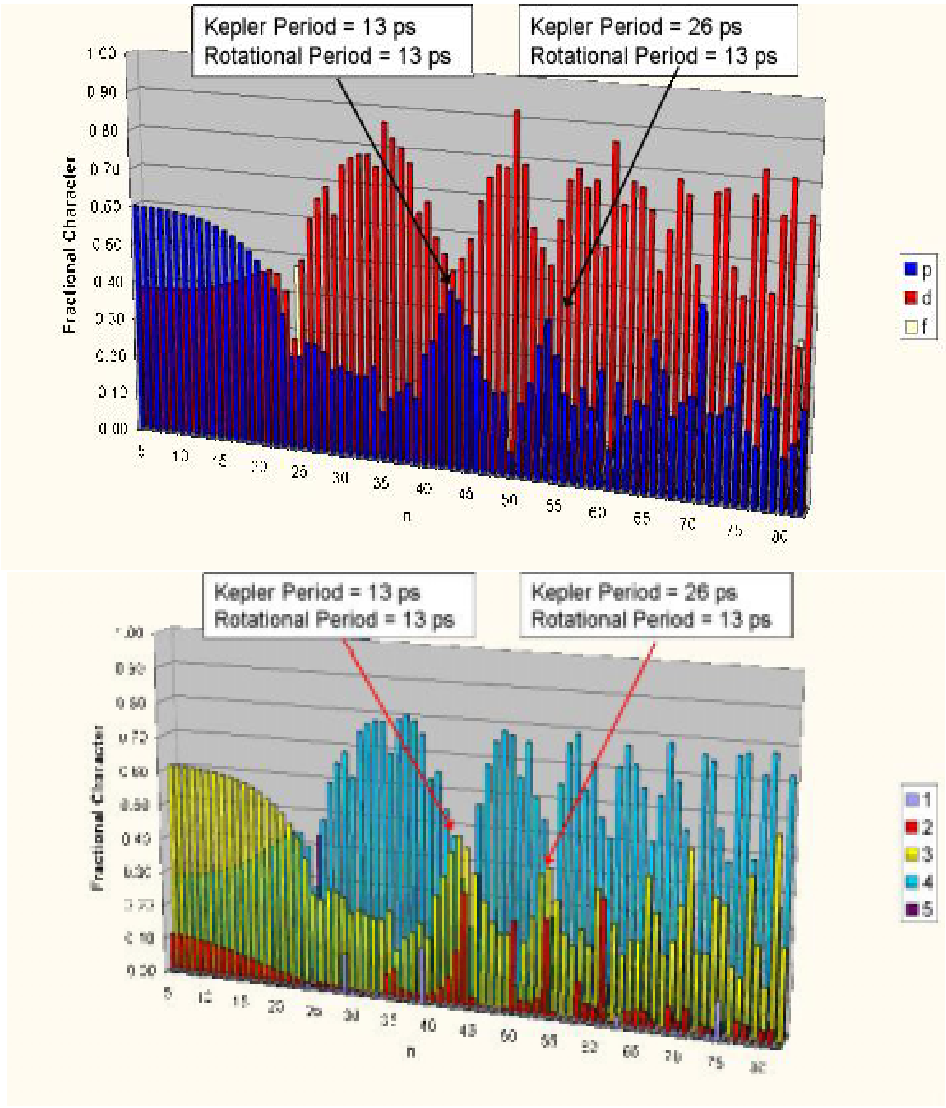

Figure 1. Composition of the pÐ series of CaF as a function of principal quantum number *n*. (a) Electron orbital decomposition. (b) Rotational decomposition. Stroboscopic resonances are indicated.

Interaction, *continued from page 5*

tance of the electron from the ion-core becomes larger, the coupling between the electron and ion should become considerably weaker. However, as the dipole moment of the CaF$^+$ core is on the order of 9 Debye, it would seem as though electronic and rotational motion would be hopelessly entangled on all time scales.

To generate a ball-and-spring mechanistic picture of how electronic and rotational motions interact in calcium monofluoride, we have used multichannel quantum defect theory (MQDT) to calculate molecular wavefunctions for each of the Rydberg states with principal quantum number *n* between 5 and 100. To determine the strength of interactions between electronic and rotational motions, we decompose the wavefunctions into their respective angular momenta. This allows us to assess the extent to which the motions of the electron and ion-core influence each other: when the wavefunctions can be described in terms of the *l* and N^+ quantum numbers, the interaction is weak; where they cannot, the interaction is strong. Representative results of the calculations are shown in Fig. 1. The figure shows the compositions of successive members of one of the six Rydberg series of CaF as a function of the principal quantum number. The decomposition in terms of orbital angular momentum is shown in (a) and in terms of rotational angular momentum in (b). As is evident in the figure, at low principal quantum number, where the electron orbits fast relative to the rotational motion of the ion-core, both *l* and N^+ poorly describe the molecular wave functions; the eigenstate wave functions have amplitude in several electronic and rotational channels. As the principal quantum number increases, *l* and N^+ become reasonably good labels: the wave functions have amplitude primarily in one electronic and one rotational channel. We thus infer that as electronic motion slows, the electron and ion begin to move independently. The large features located at *n* = 45, 55, etc. are examples of "stroboscopic" resonances and occur when the period of electron orbital motion is an integer multiple of the period of ion-core rotation. Under such circumstances, the electron sees the ion-core in the same orientation each time it approaches; the ion-core *appears stationary* from the point of view of the electron. Consequently, at such special resonances, the angular momenta of the two particles become strongly coupled just as they are at low *n*. This allows us to develop an intuitive mechanistic picture of rotational-electronic dynamics valid on all time scales: when electronic and rotational motion are in a stroboscopic relationship to each other (i.e. "resonant") the particles interact strongly through the anisotropic (axially symmetric) field of the nuclei. When the condition for the stroboscopic resonance is not satisfied, the interactions between electron and ion-core "average out": as shown in Fig. 2, each time the electron approaches the ion (once per orbit) it is in a *different* orientation. Consequently, the cumulative effects of successive electron-ion collisions cancel one another.

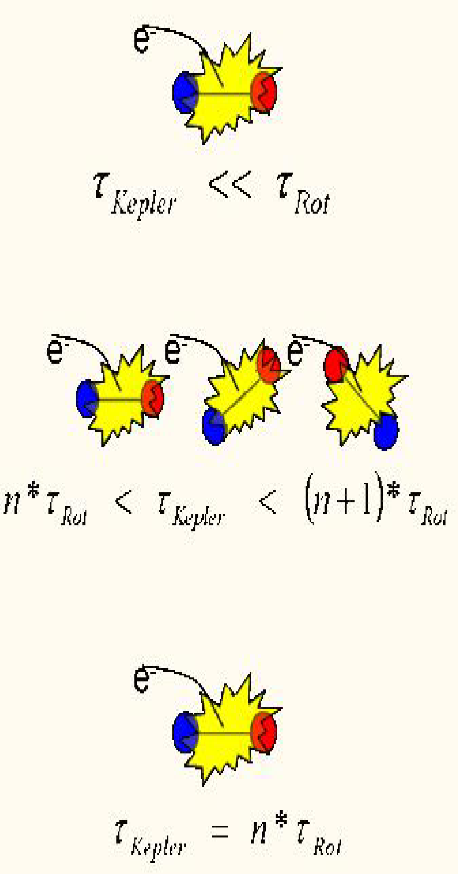

Figure 2. Artist's conception of dynamic regimes in the high Rydberg states of CaF. Relative timescales of electronic (τ_{Kepler}) and rotational (τ_{Rot}) motion are indicated below each illustration.

In addition to our study of the interaction between electronic and rotational motion described here, we are also working both theoretically and experimentally on other electron<–>nuclear energy exchange processes, especially those that lead to chemical transformations such as dissociation and ionization.

Rennovations, *continued from page 1*

structure that employed the most advanced engineering design and construction techniques then available. Great care was taken to reduce mechanical and thermal noise. The basic design called for an inner building, which housed the research laboratories, and outer building, with an entirely independent foundation separated by a six inch air gap. The inner research building was designed to float relative to the outer shell, with no connection between the two. The isolated floors of the inner building had an elaborate construction, with floors composed of three feet of reinforced concrete built upon the concrete piles. A special isolated floor covered the concrete base of the outer shell. This floor was mechanically damped using special vibration-absorbing structures built up in layers of fine sand, roofing felt transit board, vibration-absorbing cork, and concrete, and finished with an eight inch reinforced concrete floor with a smooth cement upper surface. Seismograph tests of the completed structure showed that lateral vibrations were almost completely absent and that vertical vibrations were very greatly reduced.

In order to minimize the effect of outside changes in laboratory temperatures in the building, the entire outside wall of the building was lined with an eight inch layer of cork embedded between twelve inch masonry walls. Furthermore, the large grating rooms on the first floor, where constancy of temperature was most important, were surrounded by an additional twelve inch concrete wall with a six inch air gap.

The Spectroscopy Laboratory has a distinguished 75 year history of advances in atomic physics, optics, lasers and spectroscopy. In its early years, the Laboratory became famous under Harrison's leadership for its contributions to atomic spectroscopy, including the analysis of complex spectra and qualitative and quantitative analysis of materials. This research eventually resulted in the MIT Wavelength Tables [3]. During the period from 1940 to 1946, many of the Laboratory's scientists became occupied with national defense and war work, and their spectroscopic research projects were gradually reduced. During much of this time, the Laboratory's facilities were used for spectrochemical analysis for the Manhattan Project, and many thousands of uranium samples collected from the first atomic reactor at the University of Chicago were ana-

Rennovations,*continues on page 7*

Rennovations, continued from page 6

lyzed with the help of the 10 meter grating spectrograph.

In 1946, Richard C. Lord assumed directorship of the Laboratory. Lord, an expert in infrared and Raman spectroscopy, established both of these areas in the Laboratory. Among the scientific achievements during this period were discovery and exploitation of the anomalous far-infrared spectra of ring molecules, the first vibrational analyses of the laser Raman spectra of proteins, and the important demonstration by Raman spectroscopy that the conformation of transfer RNA as determined by X-ray diffraction from a crystalline sample is the same as that in the aqueous medium existing in living cells.

Also during this period, the interests of Dr. Harrison, now Dean of the School of Science at MIT, turned to the development of spectroscopic instrumentation. In 1949, he invented the echelle spectrograph, which combines very high resolution with broad spectral range. He developed engines for the ruling of diffraction gratings under interferometric control, by which gratings of unprecedented size (as large as 400 x 600 mm) and optical performance were produced. Several of the laboratories were outfitted for stable operation of ruling engines, one provided by the Nobel Prize winning physicist A. A. Michelson.

With the arrival of Charles H. Townes and Ali Javan in the Laboratory in 1962, the possibilities of using the recently invented laser in spectroscopic research began to be explored. Townes, awarded the Nobel Prize in Physics in 1964 for his contributions in the 1950s to the development of the maser and the laser, and Javan, inventor the gas laser in 1960, made numerous achievements in the Laboratory, including the development of a version of the Michelson-Morley experiment using infrared lasers, which gave an improved upper limit on the anisotropy of space, or the effect of “ether drift,” and pioneering studies by Townes and his colleagues of the stimulated Raman and Brillouin effects. Another important achievement was the observation by Javan of the first Lamb dip and its applications to measurement of the 1.15 micron Ne^{20}-Ne^{22} isotope shift. This experiment marked the beginning of the field

Rennovations, continues on page 8

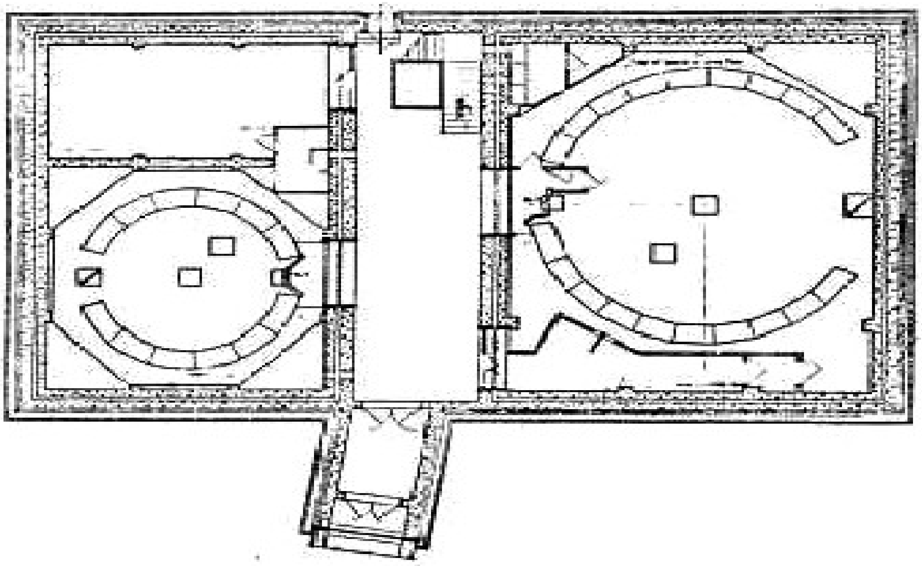

Figure 1. Original Spectroscopy Laboratory, first floor plan. From Ref.[1].

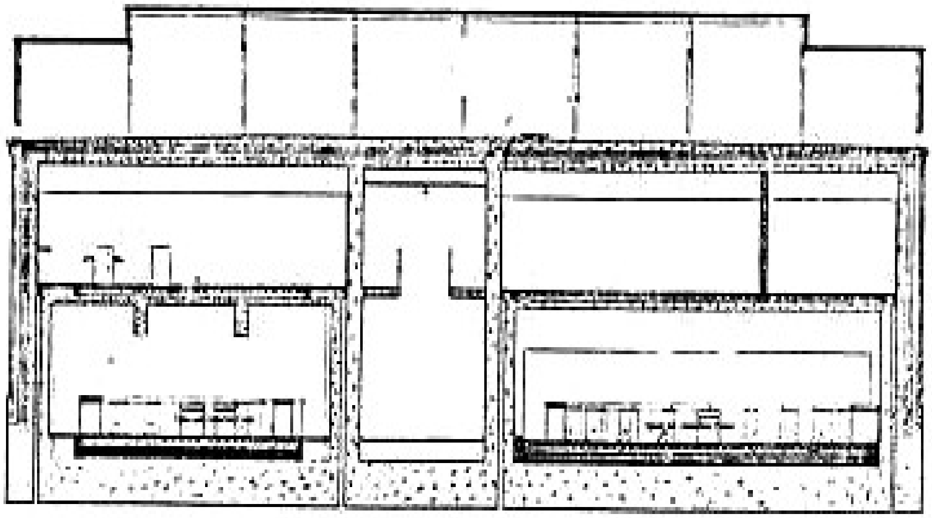

Figure 2. Original Spectroscopy Laboratory, longitudinal section. From Ref.[2].

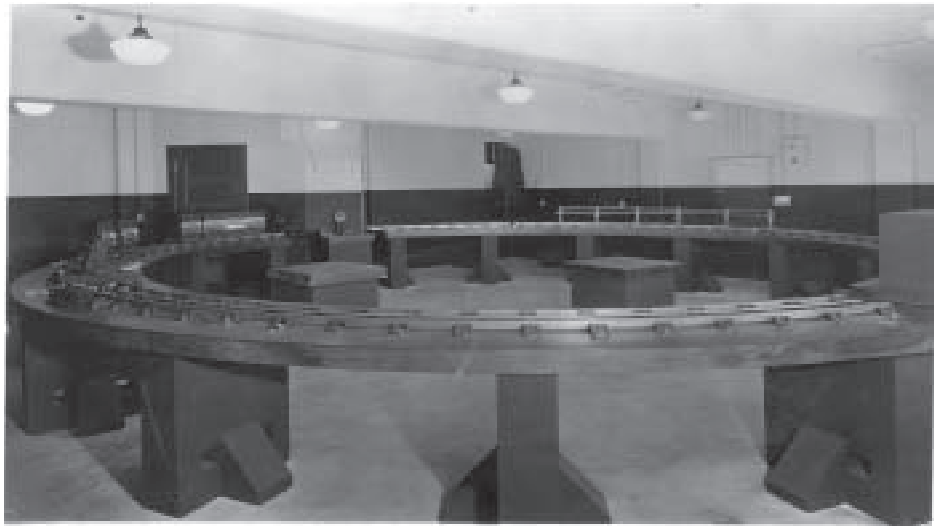

Figure 3. Ten meter walk-in spectrograph. The instrument was in the form of a Rowland circle, a spectrograph that uses a concave grating arrangement to limit light loss due to diffraction.

Figure 4. Harrison with the MIT B ruling engine. Using interferometric control, gratings of unprecedented size and optical performance were produced. The biggest, 400 mm x 600 mm, are believed to be among the largest ruled diffraction gratings ever produced. The MIT B ruling engine is still in use.

Rennovations, continued from page 7

of high-resolution laser saturation spectroscopy.

These and other studies made apparent the importance of lasers to spectroscopy and the study of dynamical processes in atoms and molecules. With the selection of Professor Michael S. Feld as Lord's successor in 1976, lasers have become a central research tool of the Laboratory. Achievements during this period include the first observation of superradiance, the discovery of a left-handed form of the DNA double helix, and the development of stimulated emission pumping as a powerful molecular probe.

Within this historic framework, the Spectroscopy Laboratory today continues to pursue fundamental and applied research in a variety of fields. Under Feld's direction two instrumentation resource centers have been established to support laser spectroscopic research in the physical and biomedical sciences. The MIT Laser Research Facility (LRF) is an NSF-supported resource for physical science research. The Laser Biomedical Research Center (LBRC), is an NIH- supported resource for research in biomedical applications of lasers, light and spectroscopy. The new physical plant will provide high quality space for the novel research directions that it faculty members are pursuing.

In the new architectural design, the foundation of Bldg. 6A will be the site of a four story structure to house the Green Center for Physics. During the period of construction, the Spectroscopy Laboratory has been relocated to temporary space in Building NW-14, the site of the MIT Magnet Laboratory.

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2. Compton, K.T., "The George Eastman Research Laboratories for Physics and Chemistry", Science 75, 472-479 (1932).

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A Fond Farewell to Recent Graduates

The following students who have been associated with the Spectroscopy Laboratory have recently obtained their Ph.D. degrees. We wish them the best of luck in their future endeavors.

Jaime Dawn Choi
Chemistry, 2005
Generation of Ultrahigh Frequency Acoustic Waves for the Characterization of Complex Materials

Inhee Chung
Chemistry, 2004
Understanding and Engineering the Photophysics of Single CdSe Nanocrystals

Joel Eaves
Chemistry, 2004
Vibrational Dynamics in Water From the molecule's Perspective

Christopher Fecko
Chemistry, 2004
Spectroscopic Investigations of Hydrogen Bond Dynamics in Liquid Water

Abigail Haka
Chemistry and Medical Physics, 2005
Development of In Vivo Raman Spectroscopy for the Diagnosis of Breast Cancer and Intra-Operative Margin Assessment

Munira Khalil
Chemistry, 2004
A Tale of Coupled Vibrations in Solution Told by Coherent Two-Dimensional Infrared Spectroscopy

Peter R. Poulin
Chemistry, 2005
Coherant Lattice and Molecular Dynamics in Ultrafast Single-Shot Spectroscopy

Nathan Stott
Chemistry, 2004
Novel Synthetic Routes to high-Quality II-VI Colloidal Nanocrystals: Controlled Growth using mild Precursors in the Presence of Selected Ligands

Joshua C. Vaughan
Chemistry, 2005
Two-Dimensional Ultrafast Pulse Shaping and its Application to Coherant Control and Spectroscopy

Mirna Jarosz Vitasovic
Chemistry, 2004
The Physics and Chemistry of Transport in CdSe Quantum Dot Solids

David W. Ward
Chemistry, 2005
Polaritonics: An Intermediate Regime Between Electronics and Photonics

Sungho Yoon
Chemistry, 2004
Model Complexes for Active Sites of Diiron Metalloproteins: Dioxygen Reactivity and Water Effects

Seminar on

Modern Optics and Spectroscopy

Spring Semester 2005

March 8	Mark Johnson, Yale University **Water at work: Using clusters to understand fundamental aspects of aqueous chemistry**
March 15	Kevin Lehmann, Princeton University **Cavity ring-down spectroscopy: From gases to cells**
March 29	Timothy Zwier, Purdue University **Single conformation vibrational spectroscopy and isomerization dynamics**
April 5	Alexander van Oudenaarden, MIT **How does a single cell remember**
April 12	Elizabeth Nolan, MIT **Small molecule fluorescent sensors for imaging zinc in biology**
April 19	Jing Kong, MIT **Orbital spectroscopy and Kondo effect carbon nanotubes**
April 26	**14th Annual Richard C. Lord Lecture** Robert Field, MIT **Just small enough**
May 3	Hrvoje Petek, University of Pittsburgh **Ultrafast imaging and control of plasma waves in Fermi seas**
May 10	Alice Ting, MIT **Fluorescent reporters of protein trafficking and function**

Tuesday, 12:00 - 1:00 p.m., Grier Room (34-401)

Refreshments served following the seminar.

Sponsored by the George R. Harrison Spectroscopy Laboratory,
Department of Electrical Engineering and Computer Science, and School of Science, MIT

Lester Wolfe Workshop in Laser Biomedicine

Optical Imaging of Pre-Cancer in the Esophagus

Tuesday, April 12, 2005, 4:00-6:00 PM
Massachusetts General Hospital
Wellman 1 Conference Room
50 Blossom Street, Boston

The incidence of cancer of the esophagus is increasing rapidly in the Western world. Chronic gastroesophageal reflux disease leads to a premalignant change in the esophageal epithelium, known as Barrett's esophagus, and this can progress to dysplasia and adenocarcinoma. The traditional method of detecting these changes employs endoscopy and taking of multiple biopsies. This workshop will discuss non-invasive optical methods of detecting and monitoring esophageal precancer, including optical coherence tomography, tri-modal spectroscopy and endoscopic confocal microscopy.

Screening and Surveillance of Barrett's Esophagus: The Challenge, Current Status and Opportunities

Norman Nishioka, Massachusetts General Hospital and Harvard Medical School

Upper Gastrointestinal Optical Coherence Tomography

Brett Bouma, Massachusetts General Hospital and Harvard Medical School

Real-Time Spectral Diagnosis of Dysplasia in Barrett's Esophagus

James Tunnell, MIT Spectroscopy Laboratory

Endoscopic Confocal Microscopy for Structure and Function

Gordon Kino, Stanford University

Refreshments served at 3:30 PM

Sponsored by the G.R. Harrison Spectroscopy Laboratory, MIT, MGH Wellman Laboratories, the Harvard-MIT Division of Health Sciences and Technology, and the Center for the Integration of Medicine and Innovative Technology

(Map available online at http://www.cimit.org/images/MGH_Map011305.pdf)

Research Article

Real-Time Spectral Diagnosis using Tri-Modal Spectroscopy

James W. Tunnell, Luis Galindo, Sasha McGee, Jelena Mirkovic, Jonathon Nazemi, Ramachandra R. Dasari, Michael S. Feld
George R. Harrison Spectroscopy Laboratory, MIT

Michael B. Wallace
Mayo Clinic, Ft. Lauderldale, FL

Christian Jost, David Lewin, Stephan Wildi
Medical University of South Carolina

Kamran Badizadegan
Massachusetts General Hospital and G. R. Harrison Spectroscopy Laboratory, MIT

The Spectroscopy Laboratory has been developing real-time spectral diagnosis, a non-invasive method for disease detection. In particular, the Laboratory has recently developed a novel spectroscopic technique, tri-modal spectroscopy or TMS, for the detection of pre-cancers in the uterine cervix,[1] esophagus,[2] and oral cavity.[3] These feasibility studies demonstrate that TMS provides a highly accurate diagnosis without the need for tissue removal. Current clinical practice in precancer detection requires guided invasive biopsy, where current guidance procedures are insufficient and biopsies are time consuming, expensive, and painful. Therefore, TMS offers an exciting opportunity to greatly improve current clinical practice by both enhancing biopsy guidance and avoiding unnecessary biopsy.

In order to leverage the advantages of spectral diagnosis, instruments and data analysis must operate in real-time (i.e. provide a diagnosis within a few seconds). We have recently reported the development of instrumentation capable of acquiring tissue reflectance and fluorescence within 1 – 3 seconds.[3, 4] Here, we describe our recent development of "real-time" TMS (< 1 s) spectroscopic algorithms capable of immediate interpretation of dysplasia and guided biopsy in patients with Barrett's esophagus (BE).

TMS combines three spectroscopic techniques (diffuse reflectance: DRS; intrinsic fluorescence: IFS; and light scattering: LSS) to provide complementary tissue morphological and biochemical parameters. DRS analyzes properties of the stromal tissue and provides the wavelength dependent reduced scattering coefficient, hemoglobin concentration and oxygen saturation.[5] IFS measures the endogenous laser induced tissue fluorescence and, using a photon migration model, corrects the fluorescence lineshape for distortions induced by tissue scattering and absorption properties.[6] Subsequently, the relative concentrations of NADH and Collagen are extracted. LSS analyzes the small portion of light (3-5%) which is singly scattering primarily from the epithelial nuclei. Analysis of this singly scattering light provides the size distribution of epithelial nuclei,[7] a known hallmark of epithelial dysplasia, a condition in which the likelihood of the tissue undergoing malignant change is greatly increased.

Eighty patients with Barrett's esophagus underwent random 4-quadrant biopsies and optical classification. A consensus diagnosis of at least 2 expert GI pathologist classified each site as non-dysplastic Barrett's (NDB), low-grade dysplasia (LGD), or high-grade dysplasia (HGD). The consensus histological diagnosis was low grade dysplasia in 23, high grade dysplasia in 15 and non-dysplasia in all other biopsies. Immediadely prior to biopsy, an optical fiber probe was brought in gentle contact with the site and sampled in less than 3 s for TMS parameters.

Figure 1 illustrates selected tissue parameters from each spectral modality. The reduced scattering coefficient decreases for HGD lesions. IFS demonstrates that the ratio of NADH to collagen increases systematically with the progression of dysplasia. Finally, LSS shows an increase in the nuclear enlargement (i.e. percent of nuclei above 10μm) for HGD lesions. A diagnostic algorithm (using logistic regression) incorpo-

TMS, continues on page 14

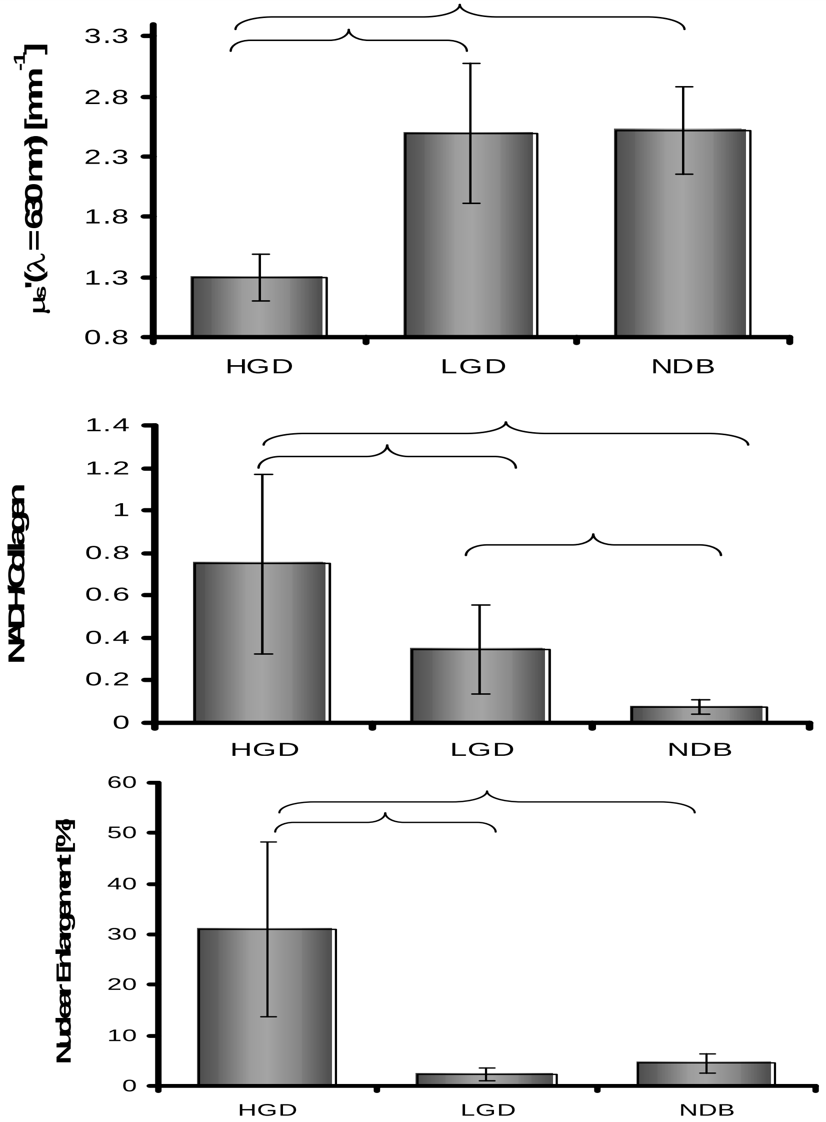

Figure 1. TMS tissue parameters extracted from (a) DRS, (b) IFS, and (c) LSS. Error bars represent 95% confidence intervals. Brackets indicate significant correlations.

TMS, continued from page 13

rating each of these parameters demonstrates the high accuracy of this technique for distinguishing HGD from all other sites (sensitivity and specifity >85%). All of these algorithms were implemented in custom software (Fig. 2), developed using National Instruments LabView and Mathworks Matlab. Analysis of the entire data set was automated and required an average of 0.8 ± 0.3 s per patient.

Real time spectral diagnosis of disease offers exciting new opportunities to aid in the management of disease detection. We have demonstrated for the first time a system capable of real time detection/analysis of HGD dysplasia in patients with BE. Real time optical detection/analysis of HGD in Barrett's esophagus using trimodal spectroscopy has the potential to guide biopsy and avoid unnecessary biopsy. Future studies aim to show that real-time guide to biopsy can enhance the detection of precancer beyond current clinical practice.

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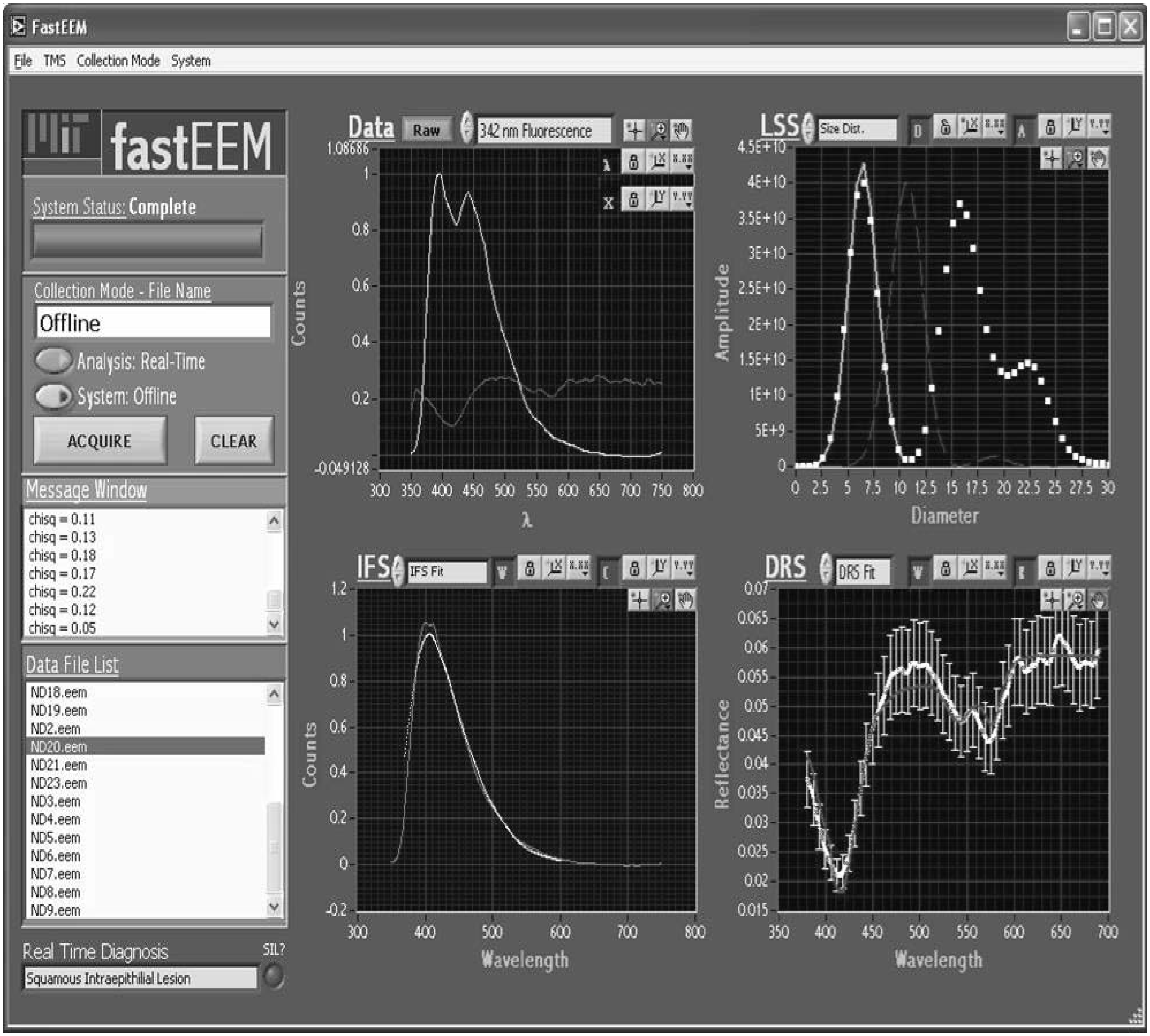

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Real Time Diagnosis: Squamous Intraepithilial Lesion

SIL?

Figure 2. Front panel of custom software designed for automated real-time diagnosis of dysplasia using tri-modal spectroscopy.

Recent Spectroscopy Laboratory Published Articles

Ahn, A., Yang, C., Wax, A., Popescu, G., Fang-Yen, C., Badizadegan, K., Dasari, R.R., and Feld, M.S., "Harmonic Phase Dispersion Microscope with a Mach-Zender Interferometer", Applied Optics 44 (7), 1188-1190 (2005).

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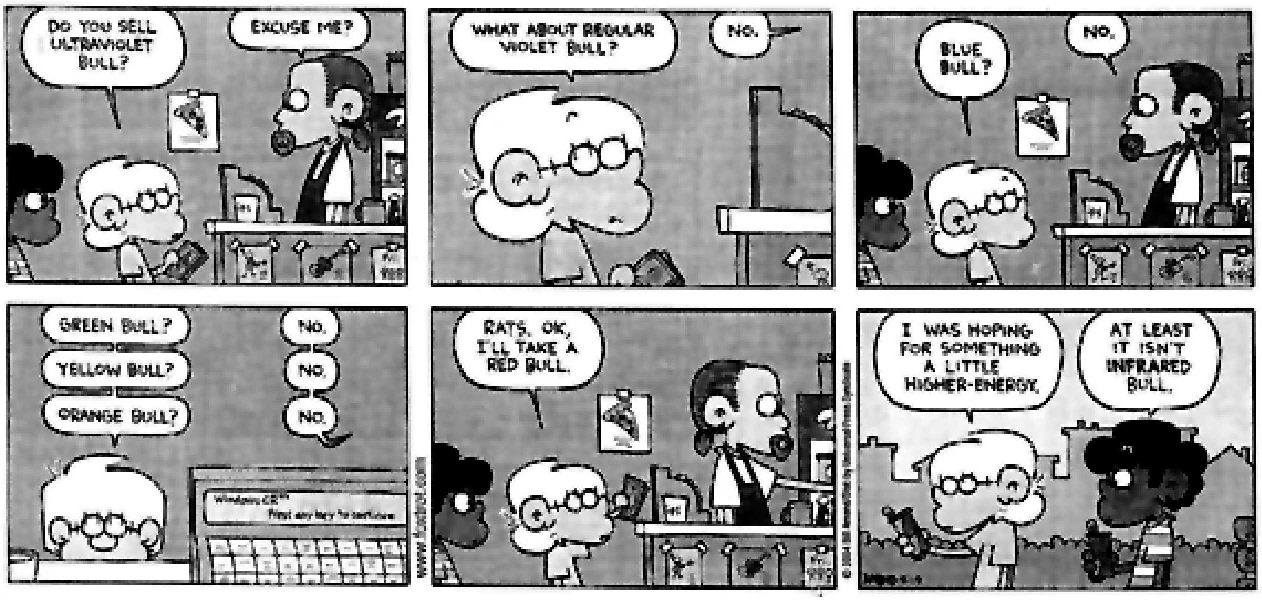

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