George R. Harrison Spectroscopy Laboratory

The George Russell Harrison Spectroscopy Laboratory conducts research in modern optics and spectroscopy to advance fundamental knowledge of atoms and molecules and explore state-of-the-art engineering and biomedical applications. The acting director is professor Robert J. Silbey. Professor Robert W. Field and Dr. Ramachandra R. Dasari are associate directors. As an interdepartmental laboratory, the spectroscopy laboratory encourages participation and collaboration among researchers in various disciplines of science and engineering. The Department of Chemistry administers personnel and fiscal matters.

Core investigators in 2010–2011 included professors Field, Moungi G. Bawendi, Keith A. Nelson, and Andrei Tokmakoff (Chemistry); professor William H. Green (Chemical Engineering); professors Mildred Dresselhaus and Jing Kong (Electrical Engineering and Computer Science); and Dr. Dasari (Spectroscopy Laboratory). The laboratory operates the Laser Biomedical Research Center, a biomedical technology resource of the National Institutes of Health, the goal of which is to develop basic scientific understanding of and technology for advanced biomedical applications of lasers, light, and spectroscopy. Core, collaborative, and outside research is conducted at the center.

Research Highlights

Professor Field, Dr. Kirill Kuyanov-Prozument, and graduate students have recorded Rydberg-Rydberg Chirped Pulse Millimeter-Wave (CPmmW) spectra in a pulsed supersonic jet by direct detection of mm-wave free induction decay signals. Dr. Prozument has constructed a pulsed, slit-jet apparatus that increases the sensitivity of the laboratory's CPmmW spectrometer by a factor >100.

Investigations continue of the acetylene S_1 state, uncovering the spectroscopic signatures of trans-cis isomerization (with professor Anthony Merer, Academia Sinica in Taiwan) and doorway-mediated intersystem crossing (with professor Wilton Virgo, Wellesley College), and of a reduced-dimension model for the trans- and cis-bent conformers (with professor John Stanton, The University of Texas at Austin).

Professor Field provided spectroscopic guidance in the use of Rydberg state vibrational autoionization for rovibronically state-selective preparation of HfF+ ions (professor Eric Cornell, University of Colorado at Boulder).

Professor Bawendi and Dr. August Dorn demonstrated nanowire/quantum dotbased photodetectors, with the dots as the light absorber and the nanowires carrying charge back to the electrodes. Professor Bawendi and his students demonstrated a low-temperature version of their novel spectroscopic method to extract fast time and high spectral information from single quantum dots. Professors Bawendi and Daniel Nocera worked with students to develop novel quantum dot probes of pH and oxygen concentration in vivo, collaborating with the Jain group at MIT. Professors Bawendi, Vladimir Bulovic, and Marc Kastner continued their studies of close-packed quantum dot films in light-emitting and photodetecting devices, demonstrating novel photodetector structures and a novel method for measuring mobilities in quantum dot films with large resistance.

Professor Nelson and his group have collected definitive proof of nondiffusive thermal transport in crystalline materials (silicon and gallium arsenide) at room temperature. Transient grating measurements with micron spatial periods over which heat transport was measured showed that the transport length did not scale with the square root of the transport time, as it would for a diffusive or random walk process, because of ballistic propagation of many of the acoustic phonons that carried that heat. This is the first measurement of room-temperature deviations from diffusive heat transport across macroscopic length scales, and it suggests that many other materials will show the same deviations. The optical generation and measurement of acoustic waves and thermal transport conducted in the Nelson lab are replicated in the Spectroscopy Laboratory's outreach laboratory for high school students, who conduct experiments on thin films and learn about advanced materials and modern optics.

Professor Tokmakoff's group investigated hydrogen bond rearrangements in liquid water, studied the dynamics of proton transfer in aqueous solution, and developed new femtosecond mid-infrared light sources. Femtosecond two-dimensional infrared spectroscopy using polarized light was used to reveal the correlated changes in hydrogen bond strength and molecular reorientation during concerted hydrogen bond switching events. These methods were also used to characterize the time scale and mechanism of proton transfer to aqueous hydroxide ion. A new ultrafast laser-driven plasma source was shown to generate mid-infrared light throughout the vibrational fingerprint region.

Professors Dresselhaus and Kong used resonant Raman spectroscopy to characterize nanocarbon materials, including single- and double-walled carbon nanotubes and single and bilayer graphene, studying these both separately and comparatively. During this past year, the Kong-Dresselhaus group continued to work as well on studies of the new, interesting electronic Raman phenomenon observed in metallic single-walled carbon nanotubes, which was discovered last year. Studies on individual triple-wall carbon nanotubes were initiated and the differences between ABA- and ABC-stacked graphene layers were also studied. Substrate effects were studied through Raman studies on a chemical vapor deposition-grown ¹³C monolayer instead of ordinary ¹²C on the Si/SiO₂ substrate, while the upper layer of bilayer graphene remained ¹²C for both samples.

The following investigators conducted basic and clinical spectroscopic biomedical studies: Drs. Dasari, Zahid Yaqoob, Timothy Hillman, Yongjin Sung, Jeon Kang, Ishan Barman, and Narahara Dingari (Spectroscopy Lab); Dr. Subra Suresh (Materials Science and Engineering); Dr. Jacquin Niles (Bioengineering); Dr. Marc Kirschner (Harvard Medical School); Dr. Kenneth Anderson (Dana Farber Cancer Institute); Dr. Wonshik Choi (Korea University), Dr. YongKeun Park (Korean Advanced Institute of Science and Technology), Dr. Maryann Fitzmaurice (University Hospitals of Cleveland), Dr. Elizabeth Stier (Boston Medical Center), and Dr. Tom Jeys (Lincoln Laboratory).

Drs. Yaqoob, Sung, and Choi continued to develop refractive index tomography

and, in collaboration with Dr. Anderson, applied it to detect myeloma cells. Dr. Yaqoob developed a new technique for observing plasma and nuclear membrane motion in eukaryotic cells, studied cell electromotility, and improved the contrast of existing quantitative phase microscopy through the use of the dispersion property of biomolecules. In collaboration with Dr. Kirschner, Drs. Sung, Dasari, and Yaqoob continued to measure cell growth using cell dry mass as the growth marker. Drs. Park, Dasari, and Suresh continued to study cell membrane fluctuations in human red blood cells to assess membrane mechanics during morphological changes.

Drs. Hillman, Park, and Yaqoob designed and implemented a new optical instrument based on digital phase conjugation to counter the effects of multiple light scattering in highly turbid media.

Drs. Barman, Dingari, and Dasari investigated advanced modeling strategies, such as band-targeted entropy minimization, to incorporate a direct causal approach for determining blood glucose levels noninvasively using Raman spectroscopy. Further, their work resulted in the design of a miniaturized Raman glucose monitor, which is poised to convert the tabletop system to a hand-held one, thereby also reducing the cost per device significantly while maintaining a high optical throughput via the incorporation of novel nonimaging optical components.

Drs. Kang and Dasari developed a system that combines phase and Raman imaging to study malaria in collaboration with Dr. Niles. Initial results highlight the promise for further investigations of more complex eukaryotic cells and their changes due to different diseases.

Drs. Fitzmaurice and Dasari continued clinical studies to diagnose breast cancer in vivo during breast needle biopsies. Drs. Lue, Kang, and Dasari worked on the development of a wide-field tissue scanner for ex vivo margin assessment of breast tissue. Drs. Stier and Dasari continued clinical studies to diagnose precancer in the uterine cervix.

Future Plans

Professor Green will convene the 7th International Conference on Chemical Kinetics at MIT from July 10 to 14, 2011, with more than 240 researchers attending. Several of the sessions will focus on spectroscopic methods for measuring reaction kinetics in combustion, in the atmosphere, and in condensed phases.

Robert W. Field Associate Director Haslam and Dewey Professor of Chemistry

Robert J. Silbey Acting Director Class of 1942 Professor of Chemistry