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Precision Measurements of Absorption and Refractive-Index Using an Atomic Candle

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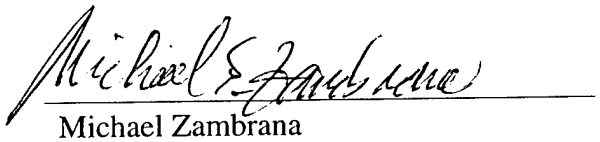

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Across a broad range of disciplines, the accurate determination of an electromagnetic wave's amplitude (either absolute or relative) has considerable relevance. Here, we demonstrate a novel and potentially very precise method for making intensity measurements based on the atomic stabilization of electromagnetic field-strength. For ease of reference, and by analogy to atomic clocks, we refer to this field-strength stabilization system as an atomic *candle*. While the candle's original purpose was to create a field with long-term intensity stability, its very nature makes it ideal for detecting subtle amplitude changes in strong electromagnetic fields, a problem that is fundamentally different from detecting weak signals in the presence of noise. In this paper, we discuss proof-of-principle experiments demonstrating the atomic candle's ability to make precise measurements of absorption coefficients and indices of refraction.

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Precision Measurements of Absorption and Refractive-Index Using an Atomic Candle

Tabitha Swan-Wood, John G. Coffer, and James C. Camparo

Abstract—**Across a broad range of disciplines, the accurate determination of an electromagnetic wave's amplitude (either absolute or relative) has considerable relevance. Here, we demonstrate a novel and potentially very precise method for making intensity measurements based on the atomic stabilization of electromagnetic field-strength. For ease of reference, and by analogy to atomic clocks, we refer to this field-strength stabilization system as an atomic *candle*. While the candle's original purpose was to create a field with long-term intensity stability, its very nature makes it ideal for detecting subtle amplitude changes in strong electromagnetic fields, a problem that is fundamentally different from detecting weak signals in the presence of noise. In this paper, we discuss proof-of-principle experiments demonstrating the atomic candle's ability to make precise measurements of absorption coefficients and indices of refraction.**

Index Terms—**Absorption coefficient, amplitude stabilization, atomic candle, atomic clocks, dielectric constant, index of refraction, magnetic resonance.**

I. Introduction

THE "SIMPLE" measurement of electromagnetic intensity is fundamental for much of experimental science. For example, in quantum optics, the QED interaction between a colored vacuum and a quantum system can be probed by examining the intensity of light transmitted through a high-Q cavity [1]. In atomic physics, the transmission of light through a resonant vapor can be used to measure atomic collision cross sections [2], while in analytical chemistry, transmitted light can reveal the presence of trace compounds [3]. New means for precisely measuring both the relative and absolute strength of an electromagnetic field, especially ones with advantageous and unique characteristics, are therefore of considerable relevance for a great many researchers.

We recently demonstrated that it is possible to actively stabilize the amplitude of an electromagnetic field to an atomic Rabi-resonance in much the same way as the frequency of a field is locked to a resonance between energy eigenstates in an atomic clock [4]. For ease of reference, and because of its analogy with the atomic clock, we refer to a field stabilized in this fashion as an "atomic candle." With regard to intensity measurements, the electromagnetic wave produced by an atomic candle has at least two unique characteristics. First, the field-amplitude of the atomic candle is essentially referenced to a dressed-atom energy level transition; hence, different candles at remote locations can produce fields with the exact same intensity. Additionally, the long-term intensity stability of the field produced by the candle has the potential for atomic-clock-like performance [5].

The atomic candle's operation derives from the response of a quantum system to a modulated field; specifically, when an atom interacts with a phase-modulated resonant field, the atomic system's population oscillates at twice the phase-modulation frequency $2\nu_m$ [6]. Of importance for the atomic candle is the fact that the amplitude of these population oscillations is a resonant function of field-strength, reaching a maximum when the Rabi frequency associated with the atomic transition matches $2\nu_m$. This resonant behavior of the oscillation's amplitude is what is meant by a Rabi-resonance, and with its observation comes an ability to lock the Rabi frequency (i.e., the field-strength) to $2\nu_m$ via an atomic signal.[1]

Our primary motivation in developing the atomic candle was to ameliorate a problem with the long-term stability of gas-cell atomic clocks [7], and to this end we constructed a microwave candle based on the ground state hyperfine transition of Rb^{87} at 6834.7 MHz [5]. Our atomic candle's microwave power stability for averaging times τ greater than 10 s is $\sigma_{\Delta P/P}(\tau) = 9 \times 10^{-7} + 10^{-7}\sqrt{\tau}$, where $\sigma^2_{\Delta P/P}$ is the Allan variance [8] of the power fluctuations relative to the Rabi-resonance peak. In order to achieve this tight lock, it is necessary for the atomic candle to detect and respond to very small changes in field intensity. Typically, these changes are stochastic in nature. However, they could just as well be deterministic and under experimental control, in which case the candle would provide a sensitive detector of subtle field-strength changes. As a proof-of-principle experiment to illustrate this potential, we have used the field of our microwave atomic candle to measure the absorption coefficient and refractive index of liquid water and acetone at 6.8 GHz.

II. Experiment

A. The Atomic Candle

Fig. 1 shows a schematic outline of our experimental arrangement, where open boxes correspond to the atomic candle proper. The heart of our candle is a resonance-cell containing isotopically pure Rb^{87} and 100 Torr of N_2 placed in the vicinity of a microwave horn. As illustrated in Fig. 2, the microwaves are resonant with the $(F = 2, m_F = 0) - (1, 0)$ ground state

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[1]Note that $2\nu_m$ may be referenced to a cesium atomic clock and hence the ground state hyperfine splitting of Cs^{133} (i.e., the SI definition of the second).

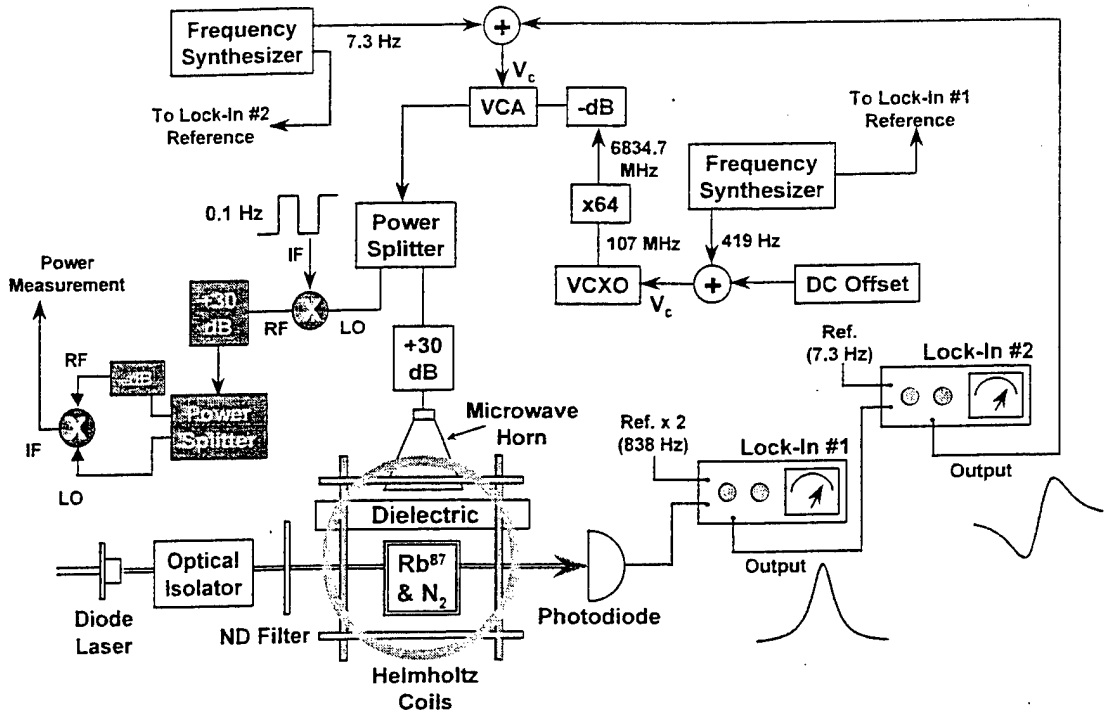

Fig. 1. Experimental arrangement. Open boxes correspond to the atomic candle proper, while filled boxes correspond to the power measurement used in the absorption measurements. LO, RF, and IF correspond to the standard local-oscillator, radio-frequency, and intermediate-frequency ports of the mixer, respectively [11].

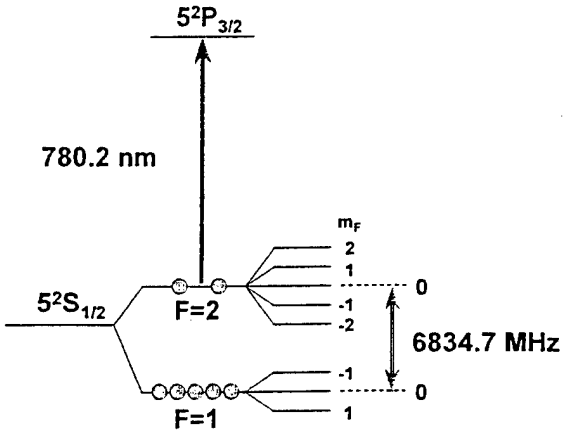

Fig. 2. Relevant energy levels of Rb^{87}. Tuning the laser to excite atoms out of the F = 2 hyperfine level, optical pumping creates a population imbalance between the $(F = 2, m_{F=0})$–(1,0) energy levels. A microwave signal resonant with the 0–0 hyperfine transition causes atoms to return to the absorbing state, thereby decreasing the light transmitted by the vapor.

hyperfine transition of Rb^{87} (i.e., the 0–0 transition). The cylindrical cell has a diameter of 2.5 cm and a length of 3.9 cm, and braided windings wrapped around the cell heat it to approximately 35 °C. The cell is centrally located in a set of three perpendicular Helmholtz coil pairs: two pairs zero out the Earth's magnetic field while the third (~300 mG) provides a quantization axis. Light from a DBR diode laser is tuned to the Rb D_2 transition at 780.2 nm [9], [19] so as to excite atoms out of the $F = 2$ ground state hyperfine sublevel. The laser beam is attenuated by neutral density filters prior to passing through the resonance cell so that the intensity entering the cell is ~60 μW/cm^2 in a beam diameter of 0.8 cm. The transmission of light through the vapor is monitored with a Si photodiode, and the propagation direction of the laser beam is parallel to the atoms' quantization axis and the microwave magnetic field.

In the absence of microwaves resonant with the 0–0 hyperfine transition, depopulation optical pumping [10] reduces the density of atoms in the $F = 2$ absorbing state, and consequently increases the amount of light transmitted through the vapor. However, when the resonant microwave signal is present, atoms return to the $F = 2$ state, thereby reducing the amount of transmitted light. The transmitted laser intensity thus acts as a measure of atomic population in the $F = 2$ level, so that any microwave-induced oscillation of this population will be observed as oscillations in the transmitted light.

The microwaves are derived from a voltage-controlled-crystal-oscillator (VCXO), whose output at ~107 MHz is multiplied up into the gigahertz regime. The microwaves are attenuated by the combination of a voltage-controlled-attenuator (VCA) and a fixed attenuator (labeled as −dB in Fig. 1) before being amplified by a +30 dB solid-state amplifier. A 419-Hz sinusoidal signal is added to a dc voltage in order to provide the VCXO's control voltage V_c. The dc level of V_c tunes the average microwave frequency to the 0–0 hyperfine resonance, while the sine wave provides microwave frequency (i.e., phase) modulation. Following the VCA, the microwave signal is split in two. One-half of the signal proceeds to the horn, which broadcasts the signal to the resonance cell, while the other half is used to directly monitor the microwave power supplied to the horn.

As already mentioned, the Rabi-resonance is manifested in the atoms' second harmonic response to the phase-modulated microwave field. The output of the photodiode is thus sent to a lock-in amplifier (labeled as #1 in Fig. 1) which is referenced to the phase-modulation's second harmonic (i.e., 838 Hz). The

photodiode/lock-in combination acts as a low-pass detector of the atoms' second harmonic signal. For the atomic candle correction signal, the microwave power is modulated at 7.3 Hz by applying a sinusoidal signal to the VCA's control voltage. The atoms' Rabi-resonance response to the modulated microwave power is monitored in a heterodyne fashion with the aid of lock-in #2, whose output becomes our field-strength correction signal. Adding this correction signal to the VCA control voltage closes the field-strength feedback control loop.

B. Measurements of Electromagnetic Field Transmission

For the absorption/refractive-index measurement portion of the experiment, the signal was chopped at 0.1 Hz prior to amplification by a second +30 dB amplifier, and following amplification the signal was again split in two. (Low-frequency chopping allowed us to discriminate against a slow baseline drift in this power-measurement portion of the experiment.) The two signals were then combined in a mixer [11], creating a dc signal whose amplitude was proportional to the power of the original microwave signal, and hence proportional to the power entering the horn.[2] If an attenuating material (in our case a dielectric liquid) is placed between the horn and the resonance cell, the microwave power reaching the Rb atoms will be reduced. As a consequence, the atomic candle will feed a correction signal back to the VCA that just compensates for this attenuation. The magnitude of the attenuated power is therefore detected as an *increase* in the microwave power supplied to the horn. Ideally, we would have used the correction signal from lock-in #2 to measure the microwave attenuation, as microwave power changes then appear on a near-zero signal background. This approach was problematic in our experiment due to limits in the dynamic range of the electronics associated with our VCA. However, we also felt that measuring the power supplied to the horn directly provides a cleaner demonstration of this atomic candle application.[3]

We employed research grade acetone and water as attenuating materials for our microwave signal. These were contained in a 14.5 cm diameter open Pyrex dish placed between the horn and the resonance cell. The dish rested on a 1.8 cm thick sheet of polyurethane microwave absorber, which had a specified microwave attenuation coefficient greater than −20 dB. The sheet was large enough to ensure that sidelobes from the horn were attenuated. A small ~3 cm^2 hole was cut in the sheet directly under the center of the horn, so that only microwaves incident normally to the liquid would pass into the resonance cell. During the course of the experiment, the acetone and water temperatures remained constant at 21 °C and 23 °C, respectively, with no observed rise in temperature due to microwave absorption. The liquid's depth d was determined by adding known volumes of liquid to the dish, and then correcting these depth values for loss due to evaporation.

[2]If V_{LO} and V_{RF} [both equal to $\sqrt{P_o}\sin(\omega t)$] are the microwave signals input to the mixer, then the mixer's output V_{IF} is just $V_{IF} = V_{LO} \cdot V_{RF} = P_o/2(1 - \cos(2\omega t))$.

[3]It might be argued that measuring the power supplied to the horn in the fashion of Fig. 1 does not really provide any advantage over more traditional techniques, as we are still making a conventional power measurement. However, one of the advantages associated with the atomic candle is its potential for very long-term stability. Hence, in the specific scheme of Fig. 1 the power measurement signal may be averaged over long time intervals to improve the measurement precision.

Subsequent to performing an absorption/refractive-index experiment, we measured the rate-of-change of power supplied to the horn dP/dt, resulting from evaporation alone.[4] At fixed temperature and pressure, the rate of acetone evaporation is constant, since this primarily depends on the acetone vapor pressure just above the liquid surface. In the case of water, the rate of evaporation is also a constant, but additionally depends on the relative humidity [12]. Consequently, by noting the times of liquid addition in the present experiment, we could use the results from these subsequent experiments to correct the relative power measurements for evaporative loss.

At microwave wavelengths, macroscopic depths of liquid act as a thin film. Therefore, the power reaching the resonance cell P_{lock} (i.e., the locked power of the atomic candle) is given by [13]

$$\frac{P_o}{P_{\text{lock}}} = \frac{e^{\alpha d}}{|T|^2}\{1 + 2|R|e^{-\alpha d}\cos[4\pi nd/\lambda + \theta] + |R|^2 e^{-2\alpha d}\}. \tag{1}$$

where

P_o	measured power supplied to the horn;
R and T	parameters associated with the reflection and transmission, respectively, of the liquid at its boundaries;
θ	phase angle;
α	absorption coefficient;
n	refractive-index.

Fig. 3 shows our acetone measurements of P_o/P_{lock} as a function of d, and these are clearly consistent with (1): damped oscillations are observed to be riding on an exponentially increasing baseline. Similar results were obtained for water, except that fewer oscillations were observed. (Though water's index of refraction at 6.8 GHz is roughly twice that of acetone's, water's absorption coefficient is more than four times larger.) Thus, in the case of water we could not access as large a range of water depths as we could for acetone due to the limitations of our microwave amplifier.

III. Results

Regarding Fig. 3, we infer α from the exponential increase in P_o/P_{lock} at large depths, while the wavelength of the sinusoidal variations in P_o/P_{lock} yields n. The results for acetone and water are given in Table I, along with estimates of the measurements' precision. Table I also presents theoretical estimates of α and n based on the empirical Cole–Cole equation, a modified version of the familiar Debye equation [14], [15]. Clearly, in its crudest realization for this type of measurement (i.e., an

[4]As an aside, we note that it should be possible to use the atomic candle to access the liquid's latent heat of vaporization. For example, in the case of acetone it is relatively straightforward to show that the rate of change of acetone depth $\dot{d}$ due to evaporation is related to acetone's latent heat of vaporization

$$\Delta Q_{vap}\,\dot{d} = B\exp\left[-\frac{\Delta Q_{\text{vap}}}{kT_{\text{liq}}}\right]$$

where B is a constant, k is Boltzmann's constant, and T_{liq} is the liquid temperature. For Beer's law absorption, where α is the absorption coefficient, $\dot{P} = -\alpha\dot{d}P_o e^{-\alpha d}$, so that if dP/dt is measured as a function of acetone temperature it should be possible to ascertain ΔQ_{vap}.

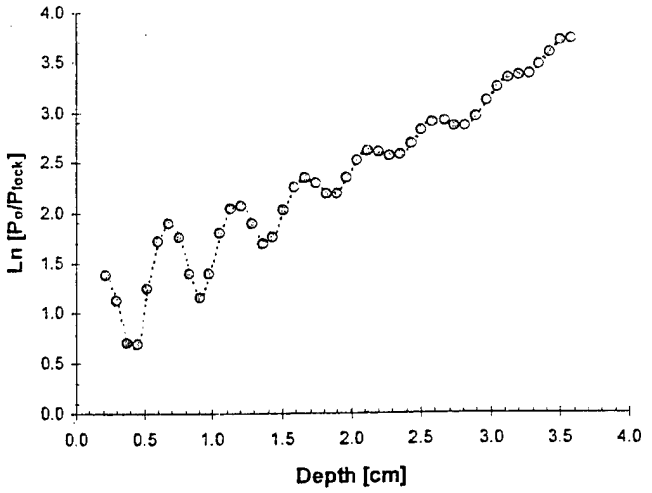

Fig. 3. Experimental results for acetone at 21 °C. Linear fit between 1.7 and 3.6 cm depths was used to determine α. After subtracting the linear slope, the oscillation wavelength yielded n. The dashed curve is simply an aid to guide the eye.

TABLE I
ABSORPTION COEFFICIENT α AND REFRACTIVE INDEX n OF LIQUID ACETONE AND WATER AT 6.8 GHz. NOTE THAT THE VALUES LISTED FOR THE EMPIRICAL COLE–COLE EQUATION ARE ESTIMATES, SINCE THE PARAMETERS EMPLOYED IN THE EQUATION ARE DEPENDENT ON TEMPERATURE AND WAVELENGTH, AND ARE THEMSELVES SUBJECT TO EXPERIMENTAL UNCERTAINTY

		α [cm^{-1}]	n
CH_3COCH_3	Atomic Candle	0.817 ± 0.026	4.623 ± 0.026
	Cole-Cole Equation	0.817	4.591
H_2O	Atomic Candle	4.07 ± 0.03	8.27 ± 0.09
	Cole-Cole Equation	4.12	8.45

open dish of evaporating liquid that attenuates a non-planewave microwave signal), the atomic candle has achieved excellent accuracy and precision in the determination of a complex dielectric constant. Moreover, the atomic candle did not need to be calibrated to a known standard [16]. For completeness, we note that it would be straightforward to construct candles based on the hyperfine transitions of Na^{23}, Rb^{85}, and Cs^{133}, so as to make dielectric measurements at 1.8, 3.0, and 9.1 GHz, respectively. It might even be possible to base an atomic candle on a Rydberg transition [17], where a much broader range of microwave frequencies would be accessible.

IV. DISCUSSION

At its most specific, this paper has demonstrated the use of an atomic candle for the precise measurement of a material's absorption coefficient and refractive index. Of course, generalizing this atomic candle application to the optical and infrared regime, one could imagine making precise cross section measurements for resonant transitions between atomic and molecular eigenstates, thereby obtaining very accurate information on the overlap of eigenstate wavefunctions.[5] However, it is our opinion that the candle's utility may extend well beyond the particular work discussed here, since we have basically demonstrated a general means for observing subtle intensity changes of an electromagnetic signal, a fundamentally different problem from that of detecting weak signals in the presence of noise.

In one possible application, a remote transmitter's output power could be detected, amplified, and made part of a local atomic candle, with the remote transmitter's output power adjusted via radio-control. Fluctuations in this atomic candle's correction signal would then provide very precise information on fluctuations in the number of scatters/absorbers along the transmitter-to-receiver propagation path. The propagation path could be terrestrial, perhaps using an optical atomic candle for environmental monitoring, or space-to-Earth. One might even envision placing two atomic candles at remote locations, with one candle transmitting a reference signal to the other. In well-known fashion, the range between the transmitter and receiver would be determined by the transmitted signal's propagation time, while Doppler shifts would provide information on the relative radial velocity. By comparing the intensity of the transmitted candle-field with the local candle-field, it might be possible to augment the range and velocity information and detect changes in the propagation geometry *transverse* to the propagation path.

It seems clear to us that with enough imagination many other atomic candle applications may be envisioned, and some of these will be much more novel than those discussed above. Consequently, we believe that the true value of the present work is not so much in describing a new means for precise absorption/refractive-index measurements, though this in itself is valuable, but rather in demonstrating the atomic candle's fundamental utility for measurement science. With the demonstration of optical and infrared atomic candles, and with a demonstration of these candles' long-term intensity stability, it is our hope that the general usefulness of atomic candles for measurement science will be realized.

[5]The microwave absorption of acetone and water in their condensed phase is properly attributed to a relaxation effect rather than resonant absorption; see [18].

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Electronics and Photonics Laboratory: Microelectronics, VLSI reliability, failure analysis, solid-state device physics, compound semiconductors, radiation effects, infrared and CCD detector devices, data storage and display technologies; lasers and electro-optics, solid state laser design, micro-optics, optical communications, and fiber optic sensors; atomic frequency standards, applied laser spectroscopy, laser chemistry, atmospheric propagation and beam control, LIDAR/LADAR remote sensing; solar cell and array testing and evaluation, battery electrochemistry, battery testing and evaluation.

Space Materials Laboratory: Evaluation and characterizations of new materials and processing techniques: metals, alloys, ceramics, polymers, thin films, and composites; development of advanced deposition processes; nondestructive evaluation, component failure analysis and reliability; structural mechanics, fracture mechanics, and stress corrosion; analysis and evaluation of materials at cryogenic and elevated temperatures; launch vehicle fluid mechanics, heat transfer and flight dynamics; aerothermodynamics; chemical and electric propulsion; environmental chemistry; combustion processes; space environment effects on materials, hardening and vulnerability assessment; contamination, thermal and structural control; lubrication and surface phenomena.

Space Science Applications Laboratory: Magnetospheric, auroral and cosmic ray physics, wave-particle interactions, magnetospheric plasma waves; atmospheric and ionospheric physics, density and composition of the upper atmosphere, remote sensing using atmospheric radiation; solar physics, infrared astronomy, infrared signature analysis; infrared surveillance, imaging, remote sensing, and hyperspectral imaging; effects of solar activity, magnetic storms and nuclear explosions on the Earth's atmosphere, ionosphere and magnetosphere; effects of electromagnetic and particulate radiations on space systems; space instrumentation, design fabrication and test; environmental chemistry, trace detection; atmospheric chemical reactions, atmospheric optics, light scattering, state-specific chemical reactions and radiative signatures of missile plumes.

Center for Microtechnology: Microelectromechanical systems (MEMS) for space applications; assessment of microtechnology space applications; laser micromachining; laser-surface physical and chemical interactions; micropropulsion; micro- and nanosatellite mission analysis; intelligent microinstruments for monitoring space and launch system environments.

Office of Spectral Applications: Multispectral and hyperspectral sensor development; data analysis and algorithm development; applications of multispectral and hyperspectral imagery to defense, civil space, commercial, and environmental missions.