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

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Prepared by

T. SWAN-WOOD, J. G. COFFER, and J. C. CAMPARO
Electronics and Photonics Laboratory
Laboratory Operations

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SPACE AND MISSILE SYSTEMS CENTER
AIR FORCE SPACE COMMAND
2430 E. El Segundo Boulevard
Los Angeles Air Force Base, CA 90245

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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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Absorption coefficient, Amplitude stabilization, Atomic candle, Atomic clocks, dielectric constant, Index of refraction, Magnetic resonance

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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.

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\(^{85}\) at 6834.7 MHz [5]. Our atomic candle’s microwave power stability for averaging times \( \tau \) greater than 10 s is \( \sigma_{\Delta P/P}(\tau) = 9 \times 10^{-7} + 10^{-7}/\sqrt{\tau} \), where \( \sigma_{\Delta P/P}^2 \) 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\(^{85}\) 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

\(^{1}\)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\textsuperscript{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\textsubscript{F} = 0, ±1) 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.

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\textsubscript{c}. The dc level of V\textsubscript{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).
photodiode/lock-in combination acts as a low-pass detector of the atoms' second harmonic signal. For the atomic carbon 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. 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.

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 18.8 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 cm² 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.

Subsequent to performing an absorption/refractive-index experiment, we measured the rate-of-change of power supplied to the horn \( \frac{dP}{dt} \), resulting from evaporation alone. 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 \( P_{\text{lock}} \) (i.e., the locked power of the atomic candle) is given by [13]

\[
\frac{P_o}{P_{\text{lock}}} = \frac{e^{\alpha d}}{|2|^2} \left( 1 + 2 |R| e^{-\alpha d} \cos[4\pi nd/\lambda + \theta] + |R|^2 e^{-2\alpha d} \right),
\]

where

- \( P_o \) measured power supplied to the horn;
- \( \alpha \) parameters associated with the reflection and transmission, respectively, of the liquid at its boundaries;
- \( \theta \) absorption coefficient;
- \( n \) refractive-index.

Fig. 3 shows our acetone measurements of \( P_o/P_{\text{lock}} \) 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 \( \alpha \) from the exponential increase in \( P_o/P_{\text{lock}} \) at large depths, while the wavelength of the sinusoidal variations in \( P_o/P_{\text{lock}} \) 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 \( \alpha \) 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

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 \( d \) due to evaporation is related to acetone's latent heat of vaporization

\[
\Delta Q_{\text{evap}} \Delta Q_{\text{evap}} d = \epsilon \exp \left( \frac{-\Delta Q_{\text{evap}}}{kT_{\text{in}}^2} \Delta Q_{\text{evap}} \right)
\]

where \( \Delta Q_{\text{evap}} \) is the constant, \( \epsilon \) is Boltzmann's constant, and \( T_{\text{in}} \) is the liquid temperature. Further calculations for the melting point of acetone, \( P_{\text{lock}} \), and the absorption coefficient, \( \alpha \), of the liquid should be possible to ascertain \( \Delta Q_{\text{evap}}. \)
Fig. 3. Experimental results for acetone at 21°C. Linear fit between 1.7 and 3.6 cm depths was used to determine \( \alpha \). After subtracting the linear slope, the oscillation wavelength yielded \( n \). The dashed curve is simply an aid to guide the eye.

### Table I

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<th>Substance</th>
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<td>( \text{CH}_3\text{COCH}_3 )</td>
<td>Atomic Candle</td>
<td>0.817 ± 0.026</td>
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<td></td>
<td>Cole-Cole Equation</td>
<td>0.817</td>
</tr>
<tr>
<td>( \text{H}_2\text{O} )</td>
<td>Atomic Candle</td>
<td>4.07 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Cole-Cole Equation</td>
<td>4.12</td>
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open dish of evaporating liquid that attenuates a non-plane wave 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\(^{25}\), 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.

### References

Tabitha Swan-Wood received the B.S. degree in physics from the University of California, Riverside (UCR), in June 2000. She is currently pursuing the Ph.D. degree in materials science from the California Institute of Technology, Pasadena.

Her research interests have included spin glass magnetism, silicon detector development, and atomic physics. She is currently studying the vibrational entropy using inelastic neutron scattering. She has three publications from her undergraduate research.

Ms. Swan-Wood was a California Robert C. Byrd Honors Scholar (1996-2000). She received UCR's Academic Program Excellence and Research Award in 1998 and 2000. She was UCR's Dr. Robert Wild Physics Scholar (1998). She received the Rosemary S. J. Schraer Outstanding Student Commencement Award for UCR's College of Natural and Agricultural Science. Upon entering California Institute of Technology, she received the Donald S. Clark Fellowship for 2000-2001.

John G. Cofer was born in Pennsylvania in 1947. He received the B.A. degree in physics from the University of Colorado, Boulder, in 1969, and the M.S. degree in physics from the University of California, Los Angeles, in 1978.

He has been Member of Technical Staff at The Aerospace Corporation, Los Angeles, since 1979. He is interested in interferometry, spectroscopy, and laser applications in applied physics and instrumentation. Recently, he has been working in the areas of diode-laser-pumped atomic clocks and fiber-optic gyroscopes.

James C. Camparo was born in Newark, NJ, in 1956. He received the B.A. degree in physics and the Ph.D. degree in chemical physics, both from Columbia University, New York, NY, in 1977 and 1981, respectively.

In 1981, he joined The Aerospace Corporation, Los Angeles, CA, as Member of Technical Staff. Currently, he is a Senior Scientist in the Lasers and Optical Physics Department of the same company. His primary interest is in the area of quantum system interactions with stochastic fields, and how the stochastic-field/atom interaction influences the operation of precise frequency standards.

His studies have included laser phase-noise to amplitude-noise conversion as mediated by resonant atomic absorption, and the role of stochastic fields in multiphoton processes. In addition to his atomic physics research, he has an interest in the design and operation of precision timekeeping systems for space applications. In support of missiles and programs, he has created computer simulations of missiles timekeeping systems, and used these to investigate the performance of spacecraft timekeeping algorithms.
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