Precision test of the ac Stark shift in a rubidium atomic vapor

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(Received 22 September 2015; published 29 February 2016)

The ac Stark shift (also known as the "light shift") is one of the most important physical processes that arises in precision spectroscopy, affecting the basic understanding of field-matter interactions, measurements of fundamental constants, and even the atomic clocks onboard GPS satellites. Although the theory of the ac Stark shift was fully developed by the 1960s and 1970s, precision tests of theory have, for the most part, been few. Taking advantage of recent developments in atomic clock technology, specifically the pulsed approach to atomic signal generation, which allows frequency measurements with a resolution of $\sim 10^{-15}$, we demonstrate a methodology for measuring the ac Stark shift. Here, we report results from a precision examination of the ac Stark shift in a vapor phase system, examining the resonant frequency of the ^{87}Rb 0-0 hyperfine transition for a perturbing laser tuned over a broad optical frequency range (18 GHz) around the D_1 absorption resonance. Over the full frequency range the agreement between semiclassical theory and experiment is very good (better than 5×10^{-2}), and in our experiments we test both the frequency dependence of the scalar and tensor components of the light shift.

DOI: 10.1103/PhysRevA.93.023433

I. INTRODUCTION

Though arising as a second-order perturbation in the field-atom interaction [1], the ac Stark shift, or "light shift," is of fundamental importance in much of basic atomic physics. It has a significant effect on multiphoton processes [2]; it plays an important role in sub-Doppler cooling for lattice traps [3], and its influence via blackbody radiation cannot be ignored in experimental realizations of the second [4,5]. Moreover, the ac Stark shift is fundamentally related to the Lamb shift. Specifically, the ac Stark shift arises from virtual transitions induced by real photons, whereas the Lamb shift arises from virtual transitions induced by virtual photons [6]. This correspondence between the ac Stark shift and the Lamb shift was already recognized by Kastler in the 1960s [7], and calculations suggest that the distinction between the ac Stark shift and the Lamb shift can begin to blur when atoms interact with fields in a colored vacuum [8].

From an applied physics perspective, one of the ac Stark shift's more significant manifestations is via the 0-0 hyperfine transition in optically pumped alkali-metal vapors, in particular the vapor-cell Rb atomic clocks [9] that fly on Global Navigation Satellite System (GNSS) spacecraft. In particular, light-shift-induced jumps can be seen (not uncommonly) in the frequency of the on-orbit Rb atomic clocks, displaying a fractional frequency magnitude $\sim 10^{-13}$ [10], and such frequency jumps are large enough to cause meter-level positioning errors.

Given the importance of the ac Stark shift from both a basic and an applied physics perspective, it is surprising that since its first observation [11] there have been few focused tests of the theory. To be clear, researchers have investigated aspects of the theory, most notably its linear dependence on light intensity [12,13]. Additionally, researchers have made comparisons between theory and experiment at specific optical wavelengths far detuned from resonance [5,14]. However, only partial and limited tests have been made on the full optical frequency dependence of the ac Stark shift near resonance, where photon scattering is strongest and dephasing processes cannot be ignored [15,16].

In one of the more detailed tests of the ac Stark shift, Arditi and Picqué tuned a diode laser over the vapor-phase D_2 resonance of Cs (*i.e.*, $6\,^2S_{1/2} \rightarrow 6\,^2P_{3/2}$ at 852.1 nm), and measured the ac Stark shift of the optically pumped Cs 0-0 ground-state hyperfine transition as a function of perturbing laser frequency (i.e., the "light-shift curve") [17]. Although the authors obtained reasonably good agreement between theory and experiment (i.e., 10% to 15%), there were a number of limitations to their work due to the measurement capabilities available in the mid-1970s. Perhaps most significantly, the laser that created the ac Stark perturbation was also the laser that optically pumped the Cs atoms. Thus, not only did the ac Stark shift vary as the laser was tuned across the D_2 resonance, but so too did the hyperfine resonance amplitude and quite likely the location within the resonance cell where the transition was generated [18–20]. Consequently, there was the very real potential of a systematic inhomogeneous light shift limiting the accuracy of their results [21]. Additionally, the light shift was assessed while the atoms were interacting with a relatively strong microwave field, and as will be discussed below we have evidence that a strong saturating microwave field can alter the form of the light-shift curve. Finally, as Arditi and Picqué note, their work was a test of only the scalar part of the light shift, since they could not resolve the $6\,^2P_{3/2}$ excited-state hyperfine splitting [22]. Although some important experimental work regarding the tensor component of the light shift was carried out in the 1960s, demonstrating its existence [23] and importantly its vanishing for isotropic light [24], it remains only poorly tested by experiment to date.

II. PRESENT THEORY AND ITS POTENTIAL LIMITATIONS

According to the semiclassical theory of the light shift [1], the ground-state evolution of an atom interacting with near-resonant light is described by an effective (non-Hermitian) perturbation operator $\mathbf{V}$. This operator acts on the atom's ground state, and can be written in terms of two Hermitian operators: $\boldsymbol{\delta\varepsilon}$, the light-shift operator, which is of primary interest in the present work, and $\boldsymbol{\delta\Gamma}$, the light-absorption operator (i.e., $\mathbf{V} = \boldsymbol{\delta\varepsilon} - i\hbar\boldsymbol{\delta\Gamma}/2$). These two operators are given by

$$\boldsymbol{\delta\varepsilon} = \frac{1}{2}(\mathbf{V}+\mathbf{V}^{+}) = -\frac{|E_o|^2}{8}(\hat{e}^*\cdot\overleftrightarrow{\boldsymbol{\alpha}}\cdot\hat{e} + \hat{e}\cdot\overleftrightarrow{\boldsymbol{\alpha}}^{+}\cdot\hat{e}^*), \tag{1a}$$

$$-i\frac{\hbar\boldsymbol{\delta\Gamma}}{2} = \frac{1}{2}(\mathbf{V}-\mathbf{V}^{+}) = -\frac{|E_o|^2}{8}(\hat{e}^*\cdot\overleftrightarrow{\boldsymbol{\alpha}}\cdot\hat{e} - \hat{e}\cdot\overleftrightarrow{\boldsymbol{\alpha}}^{+}\cdot\hat{e}^*), \tag{1b}$$

where E_o and $\hat{e}$ are the amplitude and polarization vector of the optical field, respectively [i.e., $\vec{E}(t) = E_o\hat{e}\cos(\omega t)$], and $\overleftrightarrow{\boldsymbol{\alpha}}$ is the atom's polarizability dyadic operator.

The polarizability dyadic is the fundamental atomic quantity that describes the ac Stark shift, since it carries information on the strength and symmetry of the dipole interaction (i.e., $\langle F_g,M_g|\hat{e}\cdot\vec{\mathbf{r}}|F_e,M_e\rangle$, where $|F_g,M_g\rangle$ and $|F_e,M_e\rangle$ are the angular momentum eigenstates of the atom in its ground and excited hyperfine states, respectively), as well as information on the processes that dephase the atoms' ground-excited superposition states. In present theory this latter is captured by the plasma dispersion function (or complex error function) $Z(F_g, F_e)$ [25]:

$$\overleftrightarrow{\boldsymbol{\alpha}} = \frac{\lambda e^2}{\hbar}\sqrt{\frac{2\pi^2 M}{RT}}\sum_{F_g M_g F_e}\{|F_g, M_g\rangle\langle F_g, M_g|Z(F_g, F_e)\}\left\{\sum_{\mathrm{M_e}}\vec{\mathbf{r}}|F_e, M_e\rangle\langle F_e, M_e|\vec{\mathbf{r}}\right\} \tag{2a}$$

$$Z(F_g,F_e) = \frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}\frac{e^{-u^2}du}{\{u - [[\omega_L - \omega(F_e,F_g)] + i(\frac{A}{2}+\gamma_L+\gamma_c)]]\}}. \tag{2b}$$

In Eq. (2b), ω_L is the laser frequency, and $\omega(F_e,F_g)$ is the optical resonance frequency for the transition $|F_g,M_g\rangle \rightarrow |F_e,M_e\rangle$ (where we assume that the Zeeman splittings are small, so that the optical resonance frequency depends only on the total angular momentum quantum number F). Further, A is the Einstein A coefficient for spontaneous emission, γ_L is the laser linewidth, and γ_c is the dephasing rate associated with vapor-phase collisions. The light shift of the ground-state hyperfine levels of the alkali metals, $\Delta\nu_{\mathrm{LS}}$, can therefore be written

$$\Delta\nu_{\mathrm{LS}} = -\frac{|E_o|^2}{4}\mathrm{Re}[(\langle a,0|\hat{e}^*\cdot\overleftrightarrow{\boldsymbol{\alpha}}\cdot\hat{e}|a,0\rangle - \langle b,0|\hat{e}^*\cdot\overleftrightarrow{\boldsymbol{\alpha}}\cdot\hat{e}|b,0\rangle)], \tag{3}$$

where $F_g = a = I + 1/2$ and $F_g = b = I - 1/2$ with I the alkali metal's nuclear spin.

Routinely, in order to highlight the symmetry characteristics of the atom-field interaction, the operator $\hat{e}^*\cdot\overleftrightarrow{\boldsymbol{\alpha}}\cdot\hat{e}$ is expanded in terms of spherical tensor operators $\boldsymbol{\alpha}^L$ [1], where L denotes a specific multipole moment of the interaction operator (with L taking only the values 0, 1, or 2 as a consequence of the electric dipole nature of the field-atom interaction). Briefly, there are two distinct components to the operator $\hat{e}^*\cdot\overleftrightarrow{\boldsymbol{\alpha}}\cdot\hat{e}$ with separate rotational symmetry characteristics: a laser polarization term represented by $\hat{e}^*\otimes\hat{e}$, and a component related to the angular momentum eigenstates of the atomic system $|F,M\rangle$ (both excited- and ground-state eigenvectors), which is captured within $\overleftrightarrow{\boldsymbol{\alpha}}$. The spherical tensor operators $\boldsymbol{\alpha}^L$ combine these separate components into a single operator whose rotational symmetry characteristics are captured by the parameter L: the $L = 0$ (monopole) term has spherical symmetry; the $L = 1$ (dipole) term has the symmetry of a magnetic dipole interacting with a fictitious magnetic field, and occurs only when the light field has net angular momentum along the atom's quantization axis; the $L = 2$ (quadrupole) term has the symmetry of an electric quadrupole interacting with a fictitious electric field gradient, and occurs only when the light field is nonisotropic (i.e., with regard to the field-atom interaction the light must experience a "gradient in the atom's quantization axis," so that $\Delta M = +1$, 0, and -1 transitions do not all occur with equal probability [23,24]). Consequently, the light shift can be written as a sum of terms with different overall rotational symmetry characteristics:

$$\Delta\nu_{\mathrm{LS}} = -\frac{|E_o|^2}{4}\sum_L\mathrm{Re}[(\langle a,0|\boldsymbol{\alpha}^L|a,0\rangle - \langle b,0|\boldsymbol{\alpha}^L|b,0\rangle)]. \tag{4}$$

For the present work, where we consider the ^{87}Rb atoms perturbed by a linearly polarized laser tuned to the D_1 transition, and with a polarization direction perpendicular to the atoms' quantization axis, the light shift is composed of just two terms. There is an $L = 0$ scalar light shift, $\Delta\nu_{\mathrm{LS}}^S$, and an $\mathrm{L} = 2$ tensor light shift, $\Delta\nu_{\mathrm{LS}}^T$:

$$\Delta\nu_{\mathrm{LS}}^S = -\frac{|E_o|^2G}{8}\mathrm{Re}\left\{[Z(2,1)+Z(2,2)] - \left[\frac{Z(1,1)}{3}+\frac{5Z(1,2)}{3}\right]\right\}, \tag{5a}$$

$$\Delta\nu_{\mathrm{LS}}^T = -\frac{|E_o|^2G}{8}\mathrm{Re}\left\{\frac{1}{2}[Z(2,2)-Z(2,1)] + \frac{1}{6}[Z(1,2)-Z(1,1)]\right\}, \tag{5b}$$

Here, G is a constant given by $(\lambda^2e^2f_{\mathrm{ge}}/8\pi^2m_ec^2)\sqrt{Mc^2/2RT}$ with f_{ge} the oscillator strength. For the scalar light shift, the laser polarization and Zeeman structure of the coupled states play no significant role. The scalar light shift, after accounting for the different level degeneracies, is simply the atomic level shift that arises from a sum of off-resonant virtual transitions, each associated with a different optical transition. Alternatively, the tensor light shift depends on the quadrupole symmetry characteristics of the different hyperfine optical transitions. In particular, as evidenced by Eq. (5b), the quadrupole polarizability moments have different signs (but equal magnitudes) for the transitions from a single ground-state hyperfine level to the two excited-state hyperfine levels. Thus, the tensor light shift vanishes if the excited-state hyperfine structure is not resolved, since the net quadrupole polarizability moment for the two unresolved transitions sums to zero. As a consequence, $\Delta\nu_{\mathrm{LS}}^{T}$ is the component of the light shift that would seem to be most sensitive to excited-state perturbations that may be acting on the atom, and which might lead to modifications of ac Stark shift theory depending on how those perturbations affect the atom.

Although the theory of the light shift as captured by Eq. (4) has comported itself well over the past half century, explaining light shifts in atomic clocks and magnetometers, and allowing for the generation of light-shift mitigation strategies in those devices, there are a number of issues that could call the present theory into question. Regarding Eq. (2b), use of the plasma dispersion function assumes that the stochastic nature of the field is fully captured by a simple convolution of the optical field's line shape with the atom's homogeneous absorption resonance; and then summing this convolution over the inhomogeneous Doppler shifts that occur throughout the vapor. While this is certainly appropriate for one-photon weak-field processes, the ac Stark shift by its very nature (i.e., appearing in lowest order as a second-order perturbation phenomenon) is at a minimum a two-photon process. This is worth noting, since stochastic fields can have nonintuitive influences on multiphoton processes like the Autler-Townes effect [26], which is closely related to the ac Stark shift, and enhancements in ac Stark shifts can occur if the field exhibits correlated amplitude and frequency fluctuations [27].

Regarding Eq. (2a), the theory of the polarizability dyadic assumes that the $|F_e,M_e\rangle$ are the proper eigenstates of the unperturbed Hamiltonian. However, recent experiments suggest that elastic fine-structure-mixing collisions create excited states that have both $^2P_{3/2}$ and $^2P_{1/2}$ character [28]. Thus, for some fraction of the atoms in a collision-broadened vapor, the virtual transitions are not to states $|F_e,M_e\rangle$, but to states $|\Phi_e\rangle = a|^2P_{3/2}\rangle + b|^2P_{1/2}\rangle$. Moreover, if alkali-metal–buffer-gas collisions mix fine-structure states, they most certainly mix excited *hyperfine* states, further calling into question the nature of the excited states actually coupled to the ground state through virtual transitions. Given these considerations, and the continuing ability of atomic physics to make ever more precise measurements of atomic energy level spacings, we should not be surprised at some point in the near future to find that theory and experiment are at odds. The only real question is at what level of precision the disagreement between theory and experiment will make itself manifest.

III. EXPERIMENT

Measuring the actual magnitude of the ac Stark shift and comparing those measurements to theory is experimentally problematic, and has the potential for significant systematic error. Specifically, since the magnitude of the ac Stark shift depends on the field intensity in the signal volume, one needs to accurately account for all transmission losses in the optical beam's path, which is nontrivial. Additionally, to accurately predict the magnitude of the ac Stark shift one needs to know the spatial profile of the perturbing field in the signal volume. However, from Eq. (1a) we see that $\boldsymbol{\delta\varepsilon}(\omega_L) \sim -\mathrm{Re}[\hat{e}^* \cdot \overset{\leftrightarrow}{\boldsymbol{\alpha}}(\omega_L) \cdot \hat{e}]$. Thus, rather than comparing the actual magnitude of the light shift to theory, we can test theory by examining the *form* of the light shift's dependence on the perturbing field frequency ω_L (i.e., a normalized version of the light-shift curve). In this way theory can be tested, while eliminating (or at least significantly reducing) the systematic errors that arise from estimating the perturbing field's intensity and spatial profile in the signal volume. To this end, in our work we scale theory and experiment to the measured maximum-positive and maximum-negative ac Stark shift values, and compare these normalized light-shift curves.

It is also important to note that in many previous experiments attempting to measure the light shift, hyperfine or spin polarization was created through optical pumping [29]. This presents a problem for precision tests, since the optical pumping light and the ac Stark shift perturbing field have routinely been the same [16,17], making it difficult to isolate the fundamental ac Stark shift from systematic optical pumping effects (e.g., the inhomogeneous light shift [21]). We therefore employ two lasers in our experiments. One laser, the optical pumping (OP) laser, creates a population imbalance between the ground-state hyperfine levels of ^{87}Rb. A separate laser, the perturbation laser (PL), induces the ac Stark shift of interest as it is tuned across the atom's absorption spectrum.

Moreover, light shifts have typically been accessed in cw double-resonance experiments, confounding expectations of $\boldsymbol{\delta\varepsilon}(\omega_L)$ based on a *bare-atom* Hamiltonian with measurements of $\boldsymbol{\delta\varepsilon}(\omega_L)$ in atoms subjected to a *dressed-atom* Hamiltonian [30] (e.g., in the case of the Rb ground-state hyperfine transition, atoms dressed by the microwave photons driving the 0-0 hyperfine transition). Consequently, we probe the 0-0 resonance frequency using a Ramsey separated-fields technique (temporally separated), and routinely apply the perturbing laser only during the Ramsey period when the microwave photons, as well as the optical pumping and optical detection laser photons, are absent.

Our approach exploits the pulsed optically pumped vapor-cell atomic clock as a testbed. This testbed is illustrated in Fig. 1 [31], and we note that this atomic clock has a frequency stability that should allow measurements of the ac Stark shift with a fractional frequency resolution of 10^{-15}. To briefly summarize the test setup, the OP laser [a distributed feedback(DFB) diode laser] is tuned to the D_2 absorption spectrum of Rb at 780 nm (i.e., $5\,^2S_{1/2} \rightarrow 5\,^2P_{3/2}$), and is locked to one of the sub-Doppler optical hyperfine transitions in a separate cell. During the optical pumping cycle, the OP laser creates a population imbalance between the ^{87}Rb atoms' ground-state hyperfine levels in the clock's resonance cell. The resonance cell contains isotopically enriched ^{87}Rb and a mixed N_2-Ar buffer gas with a total buffer gas pressure of approximately 26 torr; the Ar-to-N_2 pressure ratio is 1.6. The acousto-optic modulator (AOM) then switches the OP laser "off," and a microwave $\pi/2$ pulse follows, which creates a 0-0 superposition state. The superposition state

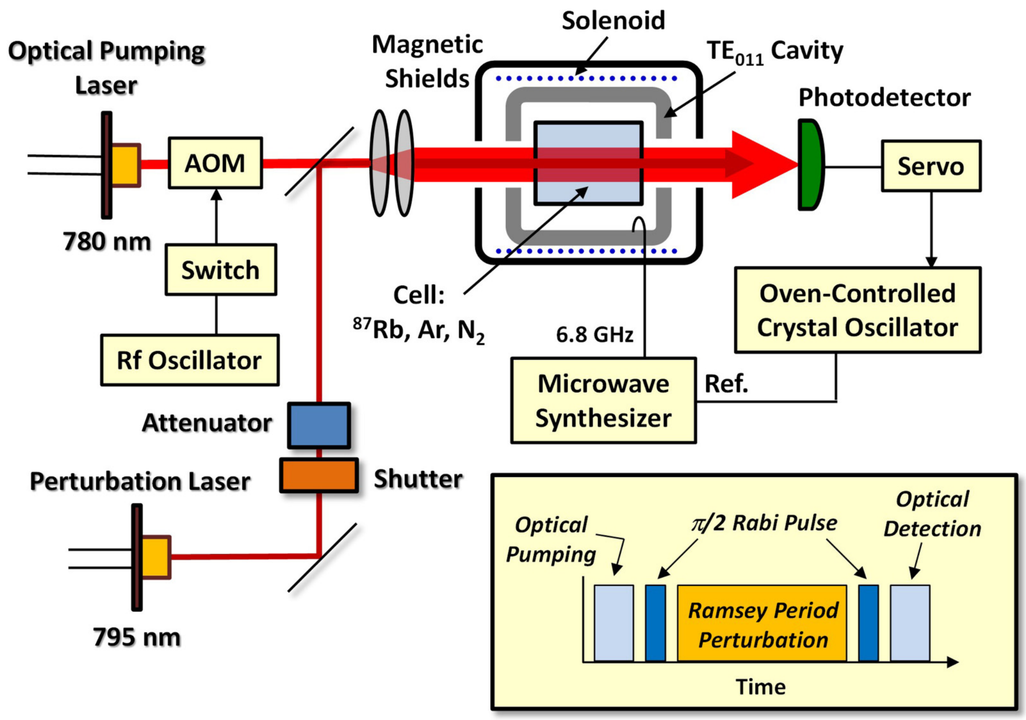

FIG. 1. Experimental setup. The pulse sequence in the box is for one (e.g., above resonance) detuning of the microwave field, and is repeated for the alternate detuning.

evolves freely during the Ramsey interval before interacting with a second $\pi/2$ pulse that returns the atoms to a bare-atom eigenstate, at which time the AOM switches the OP laser "on" with reduced intensity to detect the fraction of atoms that have made a microwave-induced state change. The frequency of the microwave pulse varies from below resonance to above resonance on alternate cycles of the sequence illustrated in Fig. 1, and the relative difference in the Ramsey signal is used to stabilize the microwave frequency to the vapor-phase atoms' 0-0 hyperfine transition resonance $\Delta\nu_{00} : 5\,^2S_{1/2}(F_g = 2, m_F = 0) \leftrightarrow 5\,^2S_{1/2}(F_g = 1, m_F = 0)$.

The perturbing DFB laser's power is kept low, so that it performs no optical pumping; it is linearly polarized in a direction perpendicular to the quantization axis of the atoms, and it has a linewidth (FWHM) of approximately 10 MHz. Consequently, tuning the PL across the D_1 transition at 795 nm (i.e., $5\,^2S_{1/2} \rightarrow 5\,^2P_{1/2}$) gives us direct access to $\mathrm{Re}[\hat{e}^* \cdot \overleftrightarrow{\alpha}(\omega_L) \cdot \hat{e}]$ over a broad frequency range without influencing the atomic clock's signal strength or the location in the resonance cell where the signal is generated. Routinely, we operate the PL in pulsed mode, and turn the PL on only during the Ramsey period. 0-0 coherence is thus an initial condition during the ac Stark perturbation. In other words, we measure the ac Stark shift of an atom perturbed by a bare-atom Hamiltonian: $H = H_o + V_{\mathrm{optical}}$, and not a (microwave field) dressed-atom Hamiltonian: $H = H_o + V_{\mu\mathrm{waves}} + V_{\mathrm{optical}}$. On alternate frequency scans of the PL over the D_1 absorption spectrum, we block the PL with a shutter for a short time in order to measure the unperturbed vapor-phase atoms' 0-0 frequency. Taking the difference yields the ac Stark shift at PL frequency $\omega_L : \Delta\nu_{\mathrm{LS}}(\omega_L) = \Delta\nu_{00}(\text{shutter open}) - \Delta\nu_{00}(\text{shutter closed})$.

The typical power density of the PL is 30 $\mu\mathrm{W/cm}^2$, and we have tested that $\pm$ 3 dB power density changes do not produce any observable change in the form of the light-shift curve, only a linear scaling of the ac Stark shift magnitude. Tuning the PL to the midpoint between the two ground-state hyperfine levels, where no hyperfine optical pumping is produced, we have verified that the light shift is linear in intensity up to PL intensities roughly 30 times higher than those employed in the work discussed here (i.e., 900 $\mu\mathrm{W/cm}^2$). Moreover, we have verified that the frequency scan of the PL is linear in time (and therefore frequency) as expected [32].

Ideally, the AOM should reduce the OP laser's nominal intensity of 20 $\mathrm{mW/cm}^2$ to zero during the Ramsey period. However, the AOM is not perfect, and 1 to 2 $\mu\mathrm{W/cm}^2$ of the OP laser light leaks into the signal volume during the Ramsey period. Consequently, if (for example) the OP laser is locked to the sub-Doppler $5\,^2S_{1/2}(F_g = 2) \rightarrow 5\,^2P_{3/2}(F_e = 1)$ transition, then during the Ramsey period there will be a slight OP laser light shift of the $|F_g = 2, M_g\rangle$ manifold of states, and hence $\Delta\nu_{00}$. Since we measure the ac Stark shift as the difference $[\Delta\nu_{00}(\text{shutter open}) - \Delta\nu_{00}(\text{shutter closed})]$, any OP laser light shift of $\Delta\nu_{00}$ should subtract out. However, as an added precaution against unforeseen systematic effects we measured our light-shift curves twice: once for the OP laser locked to a $5\,^2S_{1/2}(F_g = 1)D_2$ transition, and once for the OP laser locked to a $5\,^2S_{1/2}(F_g = 2)D_2$ transition. For the OP laser exciting atoms out of the $|F_g = 1\rangle$ state, we restricted our analysis of the ac Stark shift to PL frequencies near the $5\,^2S_{1/2}(F_g = 2)D_1$ transition (i.e., to frequencies near the *other* ground-state hyperfine level). Similarly, for the OP laser exciting atoms out of the $|F_g = 2\rangle$ state, we restricted our analysis to PL frequencies near the $5\,^2S_{1/2}(F_g = 1)D_1$

transition. The full light-shift curve was then obtained by stitching these two data sets together at a mid-point frequency.

IV. RESULTS

A. Microwave saturation and the light shift

Although we typically operate the system in the pulsed mode, with the PL applied only during the Ramsey period, it is straightforward to vary the timing of the PL so that atoms are perturbed only during the Rabi $\pi/2$ pulse. In this way, we could compare $\mathrm{Re}[\hat{e}^* \cdot \overleftrightarrow{\alpha} \cdot \hat{e}]$ for the bare-atom Hamiltonian and the dressed-atom Hamiltonian. Further, we can turn off the pulsing, and operate the system in a fully cw fashion, similar to the experiments of Arditi and Picqué [17]. An example of $\mathrm{Re}[\hat{e}^* \cdot \overleftrightarrow{\alpha} \cdot \hat{e}]$ measured under these latter conditions is shown in Fig. 2, along with the theoretical expectation of $\mathrm{Re}[\hat{e}^* \cdot \overleftrightarrow{\alpha} \cdot \hat{e}]$ for a linearly polarized field [1]. This measurement was made well into the microwave saturation regime of the 0-0 transition, where the atom's structure is defined by a dressed-atom Hamiltonian. The large theory-experiment discrepancy shown in Fig. 2 demonstrates that there are qualitative and quantitative differences in $\mathrm{Re}[\hat{e}^* \cdot \overleftrightarrow{\alpha} \cdot \hat{e}]$ for atoms subject to bare-atom and dressed-atom Hamiltonians. Though we will not pursue this topic further here, as it would lead us too far afield and is better saved for a future detailed study, we do note that Fig. 2 is disconcerting for "precision" tests of the ac Stark shift when the states are subject to an additional strong resonant field as in cw double-resonance experiments. Here, of course, we test the ac Stark shift when the additional field is absent (i.e., during the Ramsey period).

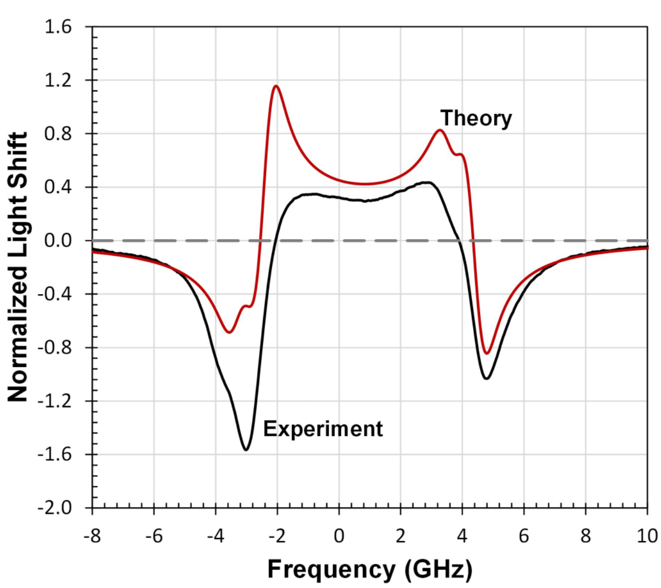

FIG. 2. Comparison of normalized light-shift curves for theory and experiment. The perturbation laser (PL) was on continuously, during both the Rabi and Ramsey periods. This is essentially the configuration for the standard double-resonance measurement procedure used to assess light shifts. In these experiments, the microwave field was well into the saturation regime of the 0-0 transition; the OP laser was tuned to the D_2 $F_g = 2$ optical resonance, and the temperature of the resonance cell was 32 °C.

B. Attenuation of the perturbation laser

As Eq. (3) indicates, the light shift will be proportional to the field intensity at the entrance of the resonance cell: $|E_o|^2$. However, the actual *measured* light shift is defined by the light intensity within the resonance cell, which varies with ω_L due to resonant absorption. Thus, Eq. (3) should be rewritten as

$$\Delta\nu_{\mathrm{LS}}(z) = -\frac{|E_o|^2 e^{-\kappa(\omega_L,z)}}{4}\mathrm{Re}\{[\langle a,0|\hat{e}^* \cdot \overleftrightarrow{\alpha}(\omega_L) \cdot \hat{e}|a,0\rangle - \langle b,0|\hat{e}^* \cdot \overleftrightarrow{\alpha}(\omega_L) \cdot \hat{e}|b,0\rangle]\}, \quad (5)$$

where $\kappa(\omega_L,z)$ is a frequency-dependent and depth- (z-) dependent attenuation coefficient, which is determined by the Rb density in the resonance cell at temperature T, and the Rb atoms' optical absorption cross section. This gives $\Delta\nu_{\mathrm{LS}}$ a depth dependence as well, so that what one actually measures is a (complicatedly weighted) average of $\Delta\nu_{\mathrm{LS}}(z)$ over the signal volume. To illustrate the significance of this effect for the present experiments, Fig. 3(a) compares the observed light-shift curve with theory for $T = 65\,^\circ\mathrm{C}$, while Fig. 3(b) provides the same comparison for $T = 40\,^\circ\mathrm{C}$. With the PL tuned near resonance, attenuation of the light intensity in the resonance cell tends to decrease the observed light shift, even though $\overleftrightarrow{\alpha}(\omega_L)$ might be relatively large. Alternatively, when the PL is detuned from resonance there is less absorption, which tends to increase the light intensity in the resonance cell and hence the light shift, although $\overleftrightarrow{\alpha}(\omega_L)$ might be relatively small. Thus, the frequency-dependent structure one observes in vapor-phase light-shift measurements arises from two sources, $\overleftrightarrow{\alpha}(\omega_L)$ and $\kappa(\omega_L,z)$, and this creates the potential for significant systematic error.

To control for this systematic effect, we measured the light shift at several relatively low resonance cell temperatures, where the vapor was optically thin. We then extrapolated each point on the light-shift curve to a temperature five degrees lower than our lowest measured temperature, or 25 °C in the present experiments. Several examples of these extrapolations are shown in Fig. 4. Not only does the extrapolation reduce the influence of $\kappa(\omega_L,z)$ on our assessments of the fundamental light-shift curve, but the standard errors of the extrapolated values provide a measure of uncertainty regarding the mitigation of this systematic effect.

C. Precision test of theory

Figure 5(a) shows our best estimate of $\Delta\nu_{\mathrm{LS}}(\omega_L)$ (considering both the scalar and tensor light-shift terms) for the perturbation laser tuned 18 GHz about the D_1 resonance of ^{87}Rb: (1) with the PL applied only during the Ramsey period of the pulse sequence, (2) by extrapolating the light-shift curves to low vapor temperature in order to mitigate systematic errors due to $\kappa(\omega_L,z)$, and (3) by stitching together the light-shift curves for the OP laser tuned to the $F_g = 1$ and $F_g = 2$ D_2 resonances to mitigate unknown OP laser effects. The difference between theory and experiment is highlighted in Fig. 5(b), and at maximum has a relative value of 5×10^{-2}. This small theory-experiment discrepancy maximizes near the absorption resonances, and is of the same order as our error in extrapolating the light-shift curves to 25 °C. Consequently,

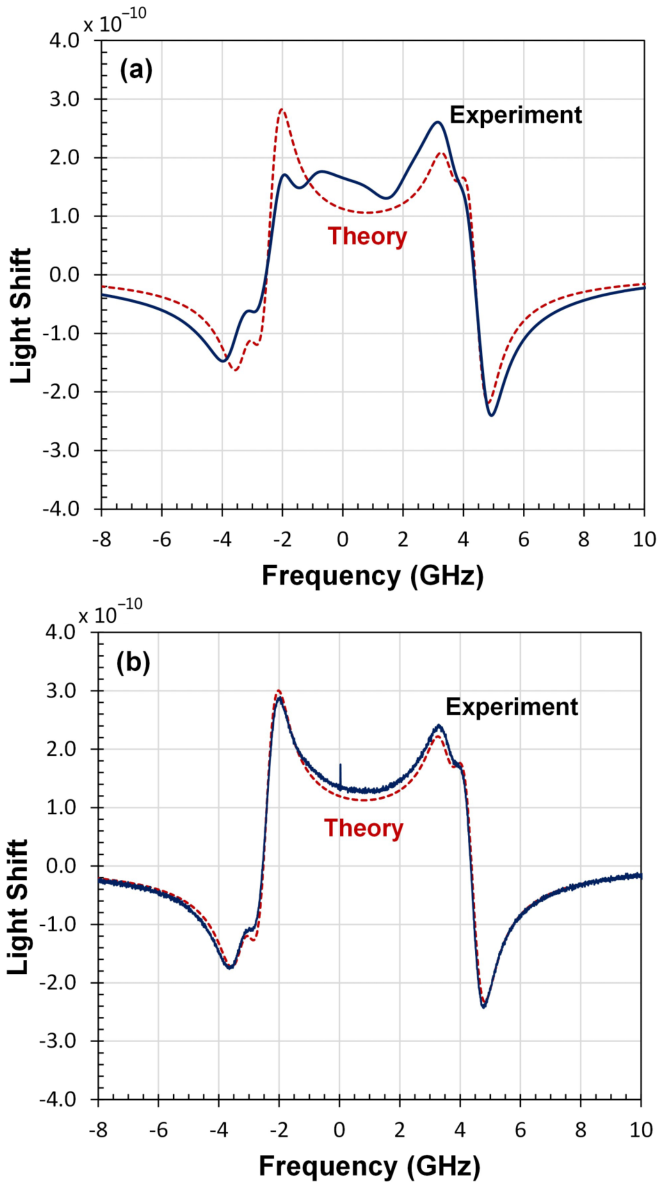

FIG. 3. (a) Comparison of the measured ac Stark shift (in units of fractional frequency) at a resonance cell temperature of 65 °C with theory; the perturbation laser is on only during the Ramsey period of the pulse sequence. (b) Same as (a) except $T = 40\,^{\circ}$C. In both cases the theory is scaled to the experimental maximum and minimum light shifts.

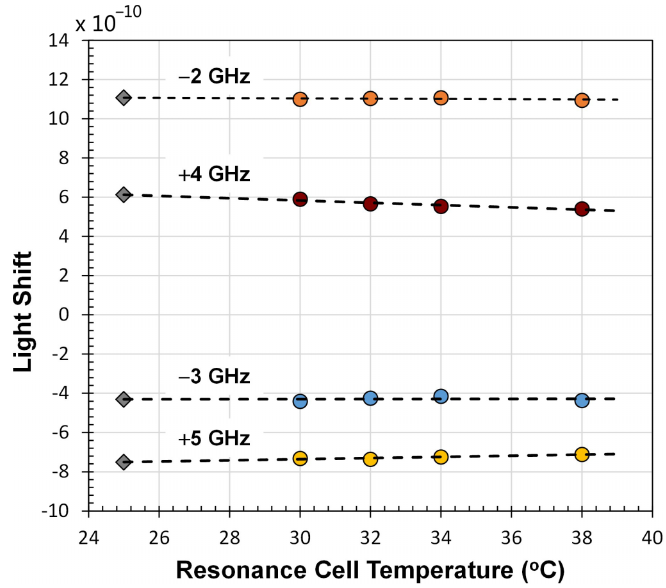

FIG. 4. Several sets of data points are plotted showing $\Delta\nu_{\mathrm{LS}}$ (in units of fractional frequency) as a function of resonance cell temperature T, for the PL detuned from resonance by the indicated amounts. We extrapolate the light-shift measurements to 25 °C, and use those extrapolated values in our comparison with theory. The near-zero slopes indicate that our values of $\kappa(\omega_L)$ at these temperatures are already quite small. Finally, we note that the standard error of the extrapolated value provides a measure of this systematic error's contribution to our final results. (The standard error is smaller than the symbol size on this graph.)

we attribute the discrepancy to the limits of our ability to mitigate the $\kappa(\omega_L,z)$ systematic effect. We note that our theory-experiment agreement improves on the previous best comparison that we know of by a factor of about 3 [17], and (as will be discussed further below) was obtained on the D_1 transition where the tensor component of the light shift cannot be ignored.

In order to generate the theoretical light-shift curve, we employed three free parameters to obtain the best fit: (1) the Lorentzian contribution to the linewidth (HWHM), $\Delta\nu_L$, used in the computation of $Z(F_g,F_e)$ (the Doppler width corresponded to a temperature of 25 °C), (2) the collision shift of the excited-state hyperfine splitting, $\partial\nu_{\mathrm{hfs}}/\partial[N_{\mathrm{BG}}]$ [28], and (3) the intensity change of the PL as the laser's injection current i_{L} was varied in order to tune the laser frequency across the D_1 resonance, $dI_{\mathrm{PL}}/d\omega_L = (dI_{\mathrm{PL}}/di_L)/(d\omega_L/di_L)$ [33]. For a nominal laser linewidth of 10 MHz and a N_2-Ar mixed buffer gas of total pressure 26 torr, we would predict $\Delta\nu_L = 443.7$ MHz [34]; for the best fit we required $\Delta\nu_L = 485.7$ MHz. The increased Lorentzian contribution to $Z(F_g,F_e)$ could be due in part to a misestimation of the total buffer gas pressure in the resonance cell (i.e., 26 torr → 27 torr), which is certainly reasonable [35], and/or an increased PL laser linewidth due (for example) to a slight amount of injection current noise (i.e., 10^{-4} fractional injection current noise could easily account for the increased value of $\Delta\nu_L$). Regardless, we take the best-fit value of $\Delta\nu_L$ as very much in line with expectations.

The collision shift of the ground-state hyperfine splitting is well known [36], and consequently we should expect a collision shift of the excited-state hyperfine splitting. For the best fit between theory and experiment we required $\partial\nu_{\mathrm{hfs}}/\partial[N_{\mathrm{BG}}] = +2.6$ GHz/amagat. Although we know of no measurements of this parameter for the ^{87}Rb $5\,^2P_{1/2}$ state perturbed by either N_2 or Ar, we note that for ^{87}Rb perturbed by Xe Driskell et al. estimated $\partial\nu_{\mathrm{hfs}}/\partial[N_{\mathrm{BG}}]$ as +0.71 GHz/amagat [28], while in the Cs-Ar system Bernabeu and Alvarez estimated $\partial\nu_{\mathrm{hfs}}/\partial[N_{\mathrm{BG}}]$ as +5.2 GHz/amagat [37]. Thus, we again find the best-fit parameter value to be quite reasonable. Finally, to obtain the best fit between theory and experiment we required $dI_{\mathrm{PL}}/d\omega_L = -0.65\%$/GHz. Not only

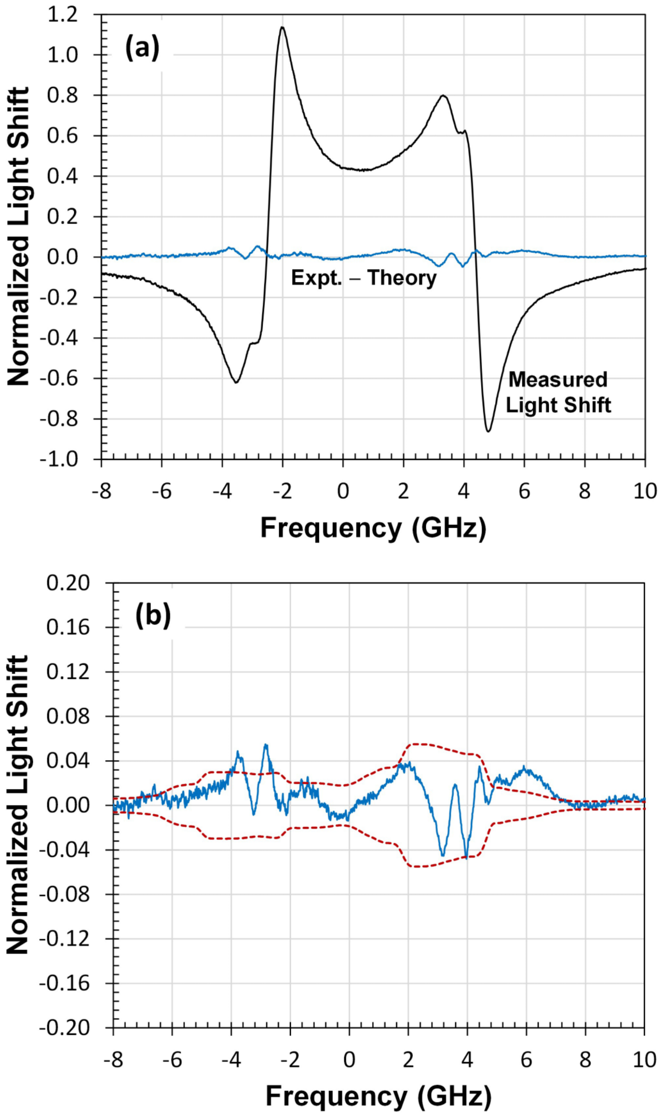

FIG. 5. (a) Measured values of $\Delta\nu_{LS}$ subject to our controls of the systematic effects: (1) perturbing laser on only during the Ramsey period, (2) extrapolation to 25 °C, and (3) stitched together light-shift curves for the OP laser tuned to $F_g = 1$ and $F_g = 2$. The difference between experiment and theory is also shown, and around resonance shows a maximum magnitude of approximately 5×10^{-2}. (b) Experiment-Theory difference on an expanded scale; the dashed lines correspond to 1σ error bounds on the extrapolated light-shift values, and suggest that the experiment-theory discrepancy is at the limits of our ability to control for the attenuation of the probe laser as it is tuned across resonance.

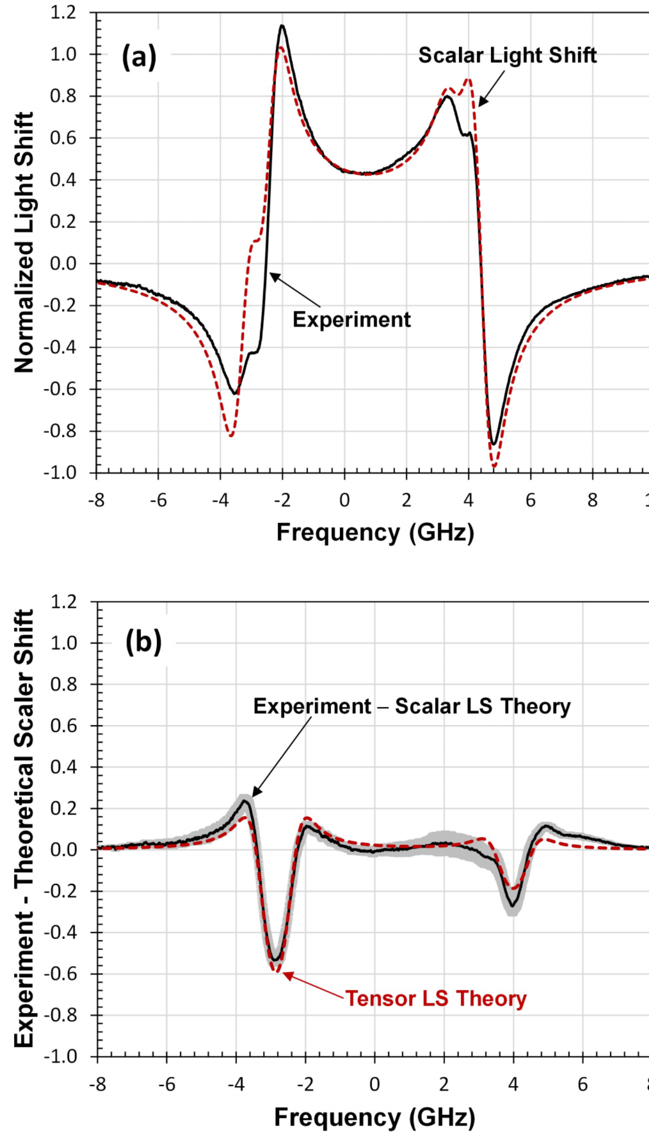

FIG. 6. (a) The experimental light-shift curve plotted with the scalar component of the theoretical light shift. The difference is clear, indicating the presence of the tensor light shift in our experiments. (b) The experimental light-shift curve after subtracting the (theoretical) scalar component is plotted with the theoretical tensor component of the light shift. The gray border around the experimental estimation of the tensor light shift corresponds to the 1σ error bars associated with our extrapolation to 25 °C.

is the sign of this parameter correct, but the magnitude is very much in line with expectations [38].

V. DISCUSSION

As noted above, the best previous test of the light-shift curve in a vapor-phase system that we know of comes from the work of Arditi and Picqué [17], who examined the ac Stark shift of the Cs 0-0 transition for a perturbing laser tuned to the D_2 transition. As a consequence, Arditi and Picqué did not resolve the excited-state hyperfine splitting, and so their experiment is solely a test of the scalar component of the light shift [i.e., $L = 0$ in Eq. (4)]. Since our perturbation laser was tuned to the D_1 transition, where the excited-state hyperfine splitting can be resolved at low enough buffer-gas pressure (such as we employ), our experiment offers a good opportunity to test the tensor component of the light shift.

Figure 6(a) shows our experimental light-shift curve again, this time plotted alongside the theoretical scalar component of the light shift. The difference between the two is clear, indicating the presence of the tensor light shift in our experimental data. To generate an experimental estimate of the tensor light shift, we subtracted the theoretical scalar light-shift curve from our experimental light-shift curve, and this is shown in Fig. 6(b). Also shown in Fig. 6(b) is the theoretical tensor component of the light shift. The gray region around the experimental estimate of the tensor light shift corresponds to the 1σ uncertainty in our extrapolation to 25 °C, and is a measure of our limits in controlling for the systematic effect of $\kappa(\omega_L,z)$. As the figure clearly shows, within the limits of that systematic uncertainty we have good agreement between experiment and theory for the tensor component of the light shift.

VI. SUMMARY

In this work we have exploited a methodology for studying the ac Stark shift that takes advantage of recent advances in atomic clock technology. Our specific experiment examined the ac Stark shift (i.e., light shift) of the ^{87}Rb ground-state 0-0 hyperfine transition in a vapor-phase system, with the rubidium atoms perturbed by an optical field tuned near the D_1 transition: $5\,^2S_{1/2} \rightarrow 5\,^2P_{1/2}$. Not only did we find very good agreement between theory and experiment over an exceptionally broad frequency range (18 GHz) about the ^{87}Rb D_1 absorption manifold (i.e., the maximum discrepancy $\cong 5 \times 10^{-2}$), but by resolving the excited-state hyperfine structure we were able to examine the tensor as well as the scalar contributions to the light shift.

In future work, we intend to examine means of decreasing the systematic contribution of $\kappa(\omega_L,z)$ to lower levels, so that ever more precise tests of the light-shift curve can be made in vapor-phase systems. As mentioned in the Introduction, collisional processes occurring in vapor-phase systems are much more complex than presently envisioned by light-shift theory, and at some level those complications will manifest themselves. Additionally, we want to test the basic theory of the ac Stark shift without such complications, and so we intend to carry these experiments forward to fountain clock testbeds. Not only will this give us an opportunity to test the semiclassical and lowest-order perturbation nature of theory, but it will be interesting to compare ac Stark shift curves for atoms perturbed by bare-atom and dressed-atom Hamiltonians. This comparison will be accomplished by measuring light-shift curves with the PL on solely during the Ramsey period, and then with the PL on solely during the Rabi period of the Fig. 1 pulse sequence. Finally, by tuning the PL to the midpoint between the ground-state hyperfine resonances, we should be able to raise the intensity of the PL to arbitrarily high levels without concern for (hyperfine) optical pumping by the PL, and in this way make precise measurements of the atom's hyperpolarizability contribution to the ac Stark shift. Regardless of the direction and breadth of these future researches, it seems clear that the methodology discussed here has much to offer our understanding of the ac Stark shift and consequently the field-matter interaction.

ACKNOWLEDGMENTS

The authors would like to thank Nathan Wells for a critical reading of the manuscript. This work has been funded by the EMRP project IND55 MClocks. EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union. The activities of J.C. were supported by The Aerospace Corporation's Sustained Experimentation and Research for Program Applications program, and funded by U.S. Air Force Space and Missile Systems Center under Contract No. FA8802-09-C-0001.

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