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Absolute absorption line-shape measurements at the shot-noise limit

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Here, we report a measurement scheme for determining an absorption profile with an accuracy imposed solely by photon shot noise. We demonstrate the power of this technique by measuring the absorption of cesium vapor with an uncertainty at the 2-ppm level. This extremely high signal-to-noise ratio allows us to directly observe the homogeneous line-shape component of the spectral profile, even in the presence of Doppler broadening, by measuring the spectral profile at a frequency detuning more than 200 natural linewidths from the line center. We then use this tool to discover an optically induced broadening process that is quite distinct from the well-known power broadening phenomenon.

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The recent development of the optical frequency comb [1,2] has delivered a powerful new tool for absorption spectroscopy. The frequency comb allows us to determine the optical frequency of spectral features to astonishing new levels, which has paved the way for intriguing laboratory-scale tests of fundamental physics [3] as well as exciting applications in a wide variety of fields, including frequency metrology [1,4], direct optical frequency comb spectroscopy [5–7], and trace gas detection [8,9]. High-quality frequency measurement, however, represents only half the story. Many spectroscopic applications also demand absorbance measurements of high precision and accuracy in order to reveal subtle features of the spectral line shape. This requirement is of particular concern to the fields of precision line-shape measurement [10,11], precision line-shape analysis [12], and Doppler thermometry [13–16].

The conventional quantum limit for precision in laser absorption spectroscopy (LAS) is set by shot noise of the probing light. In practice, though, this performance level has been exceedingly difficult to achieve because technical noise and instrumental limitations usually mask this quantum limit. These technical limitations are particularly acute for LAS because it is inherently a *bright field* [17] measurement; i.e., the absorbance is encoded in the ratio of two measurements of the incident and transmitted power. If one wishes to build a highly sensitive and accurate tool for probing optical absorbance, then there are three key challenges to overcome: First, one needs to suppress technical noise sources, which are usually much larger than fundamental sources; second, the resolution of the measurement can be limited because most of the dynamic range of the sensor is consumed in measuring small changes of relatively large signals; and third, the measurement technique needs to be inherently linear.

To overcome the technical noise and dynamic-range challenges there have been many ingenious differential-detection designs, such as the *noise eater* described in Ref. [18]. This device can be used to reach the shot-noise limit in the presence of amplitude noise up to 70 dB larger, and was recently deployed in conjunction with an optical frequency comb to achieve detection sensitivity at the shot-noise limit [19]. However, it is important to realize that taking this approach often comes at the expense of linearity, and hence accuracy. This is because either the incident power is not measured at all, or because the design of the differential detector does not ensure linearity [18,20]. This compromise is evident in the other spectroscopic techniques aimed at achieving shot-noise-limited detection such as frequency modulation (FM) spectroscopy or the extraordinarily sensitive noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) [21]. The exploitation of enhancement cavities to improve sensitivity will result in the measurement being sensitive to a mixture of absorption and dispersion [19,21]. The resulting loss in accuracy is not important if the ultimate goal is to achieve sensitivity. However, the aim of this Rapid Communication is fundamentally different: The goal is not shot-noise-limited *sensitivity* but instead shot-noise-limited *accuracy* of the absolute absorption.

The achievement of shot-noise-limited accuracy is a substantially more difficult proposition as is evidenced by the fact that careful LAS measurements to date have been a factor of ∼800 [16], ∼2500 [22], and ∼70 [23] times higher than the shot-noise limit. However, in this Rapid Communication, we demonstrate that a judicious modification to the standard LAS approach can deliver a high dynamic-range and noise-immune measurement that does not compromise the accuracy of the measurement. This enables highly accurate measurements of optical power ratios at a parts per million level, which is limited solely by shot noise on the light. We carefully measure the linearity of the measurement system and correct

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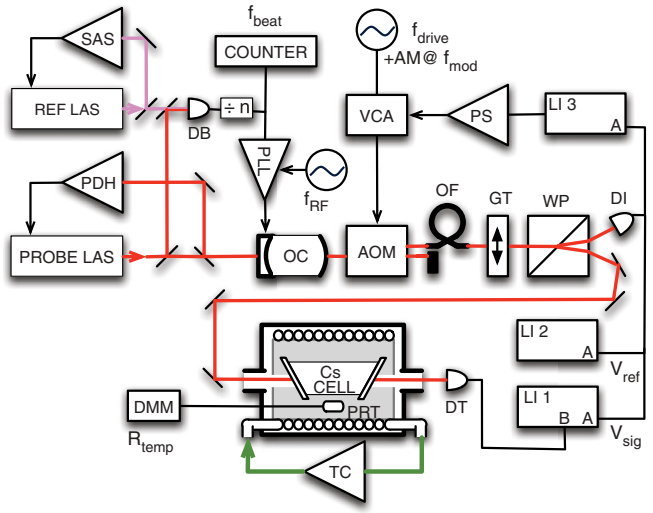

FIG. 1. (Color online) Detailed schematic of the experimental setup. An ECDL probe laser is highly stabilized and made spectrally and spatially pure. The beam is equally split into two paths: The first is detected immediately and the second is directed through a well-shielded and temperature-stabilized cesium cell and then detected. Synchronous detection of the two paths using a differential technique results in a highly linear measurement of absorbance with ppm precision.

it so that deviations from linearity are also at the ppm level. Finally, we demonstrate the power of the technique by measuring the absorption of Cs at a frequency detuning more than 400 linewidths from the D_1 transition, and then use this measurement to observe a previously unidentified optical broadening phenomena which applies to all vapor cell absorption measurements.

A detailed schematic of the experimental setup is shown in Fig. 1. An extended-cavity diode laser (ECDL), labeled *ref las*, was frequency stabilized to a saturated absorption spectroscopy (SAS) feature of cesium at 894.578 nm in the D_1 manifold, using a standard Pound-Drever-Hall (PDH) laser-locking technique [24]. A second ECDL, labeled *probe las*, was used to interrogate the transition at 894.580 nm, which is 1.17 GHz detuned from the reference transition. This laser was frequency locked using the PDH technique to the fundamental transverse mode of an optical cavity (OC) (1-MHz bandwidth; 18-GHz free spectral range). A beat note between the two lasers was formed on a high-speed photodiode (DB), whose output was frequency divided by a factor of $n = 20\,480$, and locked to a tunable radio-frequency oscillator ($f_{\rm rf}$) using a phase-locked loop (PLL) to stabilize the frequency probe at the 2-kHz level over 1–10 s. By manipulating $f_{\rm rf}$ the probe laser frequency could be tuned to an arbitrary frequency offset from the reference laser. A frequency counter was used to monitor the (divided) beat-note frequency $f_{\rm beat}$, which was used to accurately construct a frequency axis for the transition of interest.

Light transmitted by the optical cavity, which is mostly free of spontaneous emission from the ECDL, was passed through an acousto-optic modulator (AOM). This AOM performed two roles: First, it imposed an amplitude modulation on the light (90% AM depth at $f_{\rm mod} = 1.57$ kHz); and second, it acted as a variable optical attenuator to actuate a high-bandwidth power stabilization servo on the diffracted beam. The diffracted beam of the AOM was shifted in frequency by the drive frequency of 175 MHz, $f_{\rm drive}$, while the zero-order (undiffracted) beam allowed measurement of the probe laser frequency by a stabilized optical frequency comb (not shown). The diffracted beam was coupled into a length of single-mode optical fiber (OF) to eliminate pointing fluctuations, and the resulting spatially and spectroscopically pure probe beam was polarized using a Glan-Taylor (GT) prism and split equally into a reference and a signal beam with a Wollaston (WP) prism. Light from the reference beam was detected immediately, while light from the signal beam was detected after transmission through through the cesium cell.

The cesium cell under test was encased in a 15-kg cylindrical copper block. Its temperature was monitored by two platinum resistance thermometers (PRTs) calibrated by their manufacturer with an uncertainty of 30 mK. One PRT was placed at the center of the block (shown in Fig. 1), 5 mm from the midpoint of the cell; the other was located at the extreme end of the block (not shown). A temperature-controlled chiller sent refrigerated liquid through a copper pipe coiled around the block to maintain a temperature of approximately 276 K, with a stability of 3 mK over 1 min and a long-term stability of 15 mK. At this temperature the cesium vapor in the cell was optically thin, with an absorption depth of approximately 5%. The maximum gradient observed between the PRTs was 30 mK, which was at the same level as their combined calibration uncertainty. The copper block and cooling coils were enclosed within a mu-metal magnetic shield to minimize Zeeman broadening and were mounted on Sorbothane pads for passive vibration isolation.

In conventional dual-beam LAS detection, the incident and transmitted beams are independently detected to deliver a photocurrent of I and T, respectively [15,25]. Any common-mode laser intensity fluctuations are removed by constructing the ratio T/I. This procedure intrinsically compromises the resolution of an absorption measurement as most of the dynamic range of the photocurrent measurement device delivers only information on the average photocurrent. In contrast, we utilize a technique that effectively extends the dynamic range by making two separate measurements: First, a high-sensitivity measurement of the difference signal, $I - T$, which is centered on zero; and second, a separate, low-sensitivity, measurement of the reference level component (I).

The ratio T/I can then be reconstructed in software using the identity

$$\frac{T}{I} = 1 - \frac{I - T}{I}. \tag{1}$$

The increase in dynamic range of this technique over the traditional method is equal to the inverse of the depth of the absorption signal (i.e., in our case $\sim 1/0.05 = 20$). This condition is set by the maximum increase in gain allowable on the detection of $I - T$ over that of I alone.

Our implementation used two reverse-biased silicon photodiodes, with matched 47-kΩ load resistors, labeled DI and DT, to detect the incident (reference) and transmitted (signal) power, respectively. Synchronous detection of the detector outputs was performed by two lock-in amplifiers

(SRS SR830) referenced to $f_{\rm mod}$. The first (LI 1) was used to perform the high-sensitivity differential measurement by taking advantage of this device's low-noise differential input and the second (LI 2) performed the low-sensitivity reference level measurement. This careful differential approach results in an effective dynamic range sufficient to resolve optical power ratio fluctuations at the shot-noise limit.

The linearity of the entire measurement scheme (photodiode, lock-in amplifier, differencing technique) was determined using a synchronous version of the methods in Refs. [26,27]. By combining three lasers, two of nearly equal optical power and a third simulating the background level, we are able to simulate absorption features with four precisely known depths by cycling through four possible states of the equal power lasers (on:off, on:on, off:on, off:off). By variably attenuating this signal we can precisely measure the system nonlinearity (principally due to the lock-in amplifier at the level of 300 ppm [27]), and then correct the measurements to leave an estimated residual nonlinearity of 2 ppm.

Our measurements were made at probe intensities between $1.5 \times 10^{-3} I_{\rm sat}$ and $1.5 \times 10^{-1} I_{\rm sat}$ of the saturation intensity, where $I_{\rm sat} = 2.5$ mW/cm^2 [28]. At such low probe intensities, the transmitted probe power $P(f)$ can be modeled using the Beer-Lambert law [25,29],

$$P(f) = P_0(f) \exp[-\sigma], \tag{2}$$

where $P_0(f)$ is the probe power incident on the atoms at each optical frequency f and σ is the (frequency-dependent) optical depth. In this same low intensity limit, we can write $\sigma = \alpha V(f - f_0)$, where α is the on-resonance optical depth, f_0 is the optical frequency of the transition, and $V(f)$ is a Voigt function [10,25] that describes the real part of the complex susceptibility of the optical resonance for a thermal ensemble of atoms. The Voigt function is a convolution of a Lorentzian component of the line shape (homogeneous linewidth) and a Gaussian component (inhomogeneous Doppler broadening). The Lorentzian component can be defined as $L(f) = [1 + (f/\Gamma)^2]^{-1}$, where Γ is the half width at half maximum (HWHM), while the Gaussian component, $G(f) = \exp(-f/\delta_w)$, has a width of $\delta_w = \sqrt{(2kT_{\rm Cs})/(m_{\rm Cs}c^2)}$ where k is the Boltzmann constant, $T_{\rm Cs}$ is the temperature of the cesium vapor, $m_{\rm Cs}$ is the atomic mass of cesium, and c is the speed of light. Line-shape broadening associated with the linewidth of the probe laser (<1 kHz) is negligible in this experiment, while conventional power broadening amounts to just a 5% change in the homogeneous linewidth over the range of probe intensities [29]. We thus expect the Lorentzian component to be nearly constant with a width set by the natural lifetime of the transition, i.e., $\Gamma = 2.29$ MHz. All of our measurements were made at $T_{\rm Cs} = 279$ K so that all of the recorded spectra also have a fixed Gaussian component with the same characteristic width (∼207 MHz).

A typical transmission spectrum (T/I) for which $I = 0.15 I_{\rm sat}$ and $T_{\rm Cs} = 279.29 \pm 0.015$ K is shown in the lower panel of Fig. 2. A least-squares fit was performed to the spectrum and the residuals from a fit to the Eq. (2) model are shown in the upper panel of Fig. 2. The residuals are featureless and have a white spectrum with a root-mean-square (rms) spread of 2 ppm. This is entirely consistent with the expected photon shot noise at this probe power (6 μW). In addition, we held the laser at a fixed frequency and monitored the optical transmission noise as a function of time. For any given frequency the transmission noise averages as the inverse of the square root of the measurement integration time, as expected for shot noise.

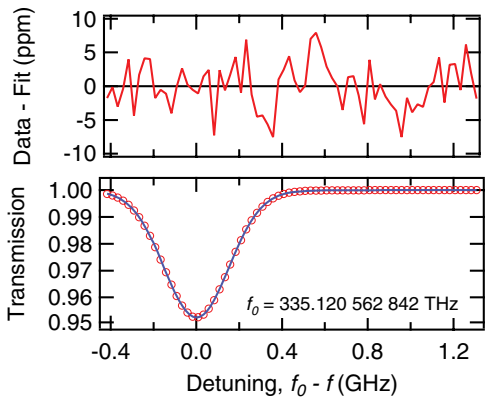

FIG. 2. (Color online) Lower panel: A typical spectrum of a D_1 Cs absorption line. Open circles are the data, and the solid blue curve is the fit. Upper panel: Residuals to the fit function in Eq. (2), which are consistent with a shot-noise-limited measurement.

Figure 3 shows the optical transmission measurement noise at 40-ms integration time as a function of probe power. Here, we see a distinct change in slope around 3.5-μW incident power from $1/P$ to $1/\sqrt{P}$ behavior. We also display the calculated Johnson noise of the photodetector readout resistor thin blue and the calculated shot noise of the light (thick green). It can be seen that the power dependence and magnitude of the noise is consistent with Johnson noise at low powers, and consistent with photon shot noise at optical powers higher than 3.5 μW.

The large dynamic range and high signal-to-noise ratio (SNR) of our approach means that we can directly reveal subtle and important changes in the absorption spectra. Figure 4 shows the frequency-dependent optical depth σ

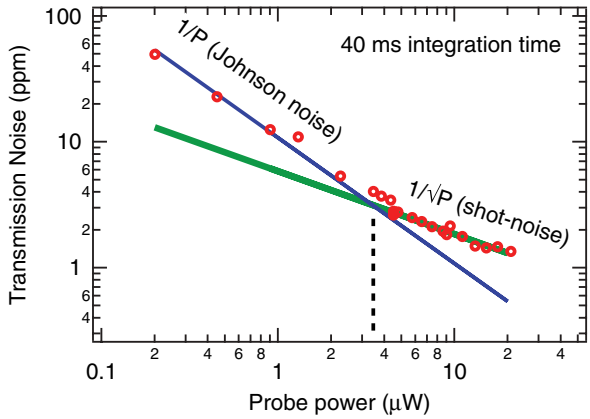

FIG. 3. (Color online) Noise in the measurement of optical transmission relative to the off-resonance value of near unity as a function of optical power for an integration time of 40 ms. The measurements become shot-noise limited at probe powers greater than 3 μW.

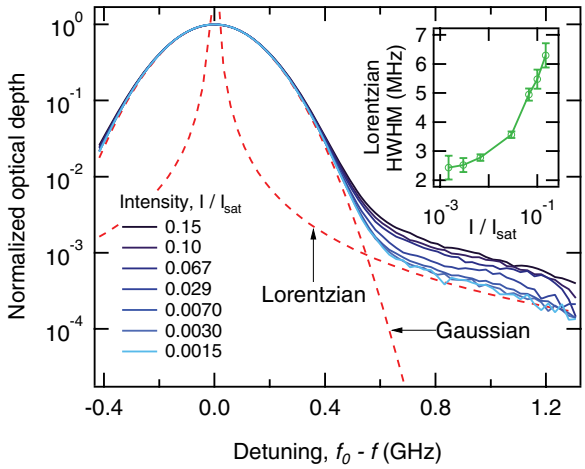

FIG. 4. (Color online) Demonstration of the breakdown in the Voigt profile at low intensities. Optical depth vs detuning for various probe intensities. The inset shows the fitted Lorentzian width as a function of probe power; the fact that this is not constant over the range of probe intensities is a signature of the breakdown of the Voigt spectral line shape [31]. The reference profile (dashed), which is expected in the limit of zero probe intensity, consists of a Gaussian component of 207.902 MHz and a Lorentzian component of 2.2875 MHz.

extracted from the data using Eq. (2). We display this depth for a set of probe powers ranging from $\sim 10^{-3}$ to $10^{-1} I_{\rm sat}$. One observes a strong on-resonance Gaussian profile associated with Doppler broadening of the vapor. In addition, the high signal-to-noise ratio in our experiment enables us to observe the Lorentzian wings that make up the "pedestal" of the Voigt profile at large detunings, $\gtrsim 100\Gamma$. These are easily distinguished: The Gaussian component decays rapidly at high-frequency detunings so that the Lorentzian wings eventually become dominant [30]. Figure 4 shows an unexpected feature: It can be clearly seen that the magnitude of the Lorentzian component is dependent on the probe intensity. This is even more evident if we replot the data in terms of its apparent Lorentzian width Γ (see the inset of Fig. 4), which increases from 2 to 6 MHz as the probe intensity increases from $10^{-3} I_{\rm sat}$ to $10^{-1} I_{\rm sat}$, whereas, from conventional considerations, we should expect this to be constant within 5% over this intensity range [28]. We have recently demonstrated that this type of behavior can arise out of a complex interplay between optical pumping and atom dynamics as an atom traverses the probe beam [31]. This interaction modifies the underlying Lorentzian line shape to give an overall line shape that can still be close to a Voigt function but with a Lorentzian width parameter that no longer equals that expected from the homogeneous processes alone.

In this work we demonstrate an approach to laser absorption spectroscopy that allows a measurement of the *absolute absorption* with shot-noise-limited accuracy. Accuracy is ensured by using a high-precision differential measurement scheme that preserves the scale information. This has permitted the reconstruction of cesium absorption spectra with a SNR limited only by shot noise on the probe light (2 ppm at 6 μW in a 40-ms bandwidth). By varying the probe power between approximately 1/100 to 1/10 of the saturation intensity, we have observed an apparent trebling of the underlying homogeneous linewidth of the D_1 transition. This observation emphasizes the power of this technique as we are making precision measurements of the homogeneous linewidth component in the wings of the spectral line more than 400 natural linewidths away from the line center. This work demonstrates that it is possible to work at the shot-noise limit for a spectroscopically accurate measurement and we would anticipate that this technique will be widely used in future spectroscopic applications.

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