Measurement of the $^{20,22}$Ne $^3P_2-^3D_3$ transition isotope shift using a single, phase-modulated laser beam

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Received 7 December 2016, revised 18 January 2017
Accepted for publication 26 January 2017
Published 10 February 2017

Abstract
We develop a simple technique to accurately measure frequency differences between far-lying resonances in a spectroscopy signal using a single, unlocked laser. This technique was used to measure the isotope shift (IS) of the cooling transition of metastable neon. Our result of $1626.287(53)$ MHz is the most accurate determination of this value to date.

Keywords: isotope shift, modulation spectroscopy, saturated absorption, sideband spectroscopy

1. Introduction
Precise measurements of atomic optical transitions usually requires overcoming the large—typically few GHz—Doppler broadening of lines caused by the thermal distribution of atoms. To this end there are a multitude of experimental techniques relying on either sample cooling (and/or trapping), or limiting the interaction with probing fields to a specific, narrow velocity group. The latter method is generally called Doppler-free spectroscopy (DFS) [1, 2], and results in narrow lines, typically a few MHz for optical transitions, which can be probed with a narrowband laser beam. Meanwhile, atomic beams or trap setups require an elaborate vacuum system and sensitive detection for small observed signals, and DFS of a thermal sample can be conducted with a gas sample in a cell, and enjoys a large signal-to-noise ratio. Finally, the systematic uncertainties in a vapor cell configuration are inherently different from those of cold atoms [3, 4].

An accurate determination of the width of and interval between atomic resonances requires calibration of the laser wavelength within the scanning range [5]. A common way to achieve this is by using a cavity with a known free-spectral-range (FSR) [6], which adds frequency markers in the form of narrow resonances whenever the laser is scanned over it. This method is limited by uncertainty and drifts in the FSR, mostly due to thermal changes in cavity length, and scan linearity. To account for nonlinearity in the scanning procedure, many close markers are desired [7, 8], which requires long cavities that are more susceptible to thermal drifts. Moreover, since the functional form of the nonlinearity is generally unknown and may change over time, interpolation errors may occur, which can be difficult to evaluate precisely.

A more elaborate method of calibrating the wavelength is to phase-lock a scan laser to a reference laser locked to a stable feature, and measure their frequency difference. This method is limited by the lasers’ noise, and the stability of the reference laser frequency during a measurement sequence. Higher stability is obtained when locking both lasers to a frequency comb [9], which is at the expense of a more elaborate and involved system.

Here we present a simple, versatile measurement scheme for the precise determination of frequency differences between far-lying resonances with different sizes. Our method overcomes most calibration challenges and drift errors, while using a single, unlocked laser. We demonstrate its applicability by measuring the isotope shift (IS) of the $2p^53s\,^3P_2$ ($134041.8400$ cm$^{-1}$) to $2p^53p\,^3D_3$ ($149657.0393$ cm$^{-1}$) transition between $^{20}$Ne and $^{22}$Ne. A closed and isolated transition used for laser-cooling applications [10].

2. Atomic signal with phase modulation
We implement DFS by saturated absorption (SA) [11]. For a single transition with a homogeneously broadened linewidth
\[ I[\Delta] = I_0 e^{-G_1(1-S_1(\Delta, T))}, \]

(1)

with \( G \) denoting the Doppler-broadened Gaussian absorption coefficient of the atomic vapor, including the atomic density and cell length; \( S \) is the resonance depth, which depends on the pump and probe intensities; and \( \Delta \) is the detuning from resonance. \( L \) is a normalized Lorentzian transmission function \( L[\Delta, \Gamma] = 1/(1 + 4(\Delta/\Gamma)^2) \). For a sample containing two isotopes with an IS of \( \omega_{IS} \), the transmission is given by

\[ I[\Delta] = I_0 e^{-G_1(1-S_1(\Delta, T))}, \]

(2)

where, assuming that the transition in both isotopes has similar linewidth, \( L_{\Delta}[\Delta] = L_{\Delta}[\Delta - \omega_{IS}] = L_{\Delta - \omega_{IS}} \), \( G_{\Delta}[\Delta] = G_{\Delta - \omega_{IS}} \), \( n_1/n_2 \) are the isotopic atomic densities. We expand (2) in \( G_S \) to get

\[ I[\Delta] = I_0 e^{-G_1(1 + G_1 S_1 L_1 + G_2 S_2 L_2 + O((G_S)^2))}. \]

(3)

A trace of a broad frequency scan of the SA signal (without subtraction), fitted with (3), is shown in figure 1. When the pump beam is amplitude modulated with a frequency \( \omega_m \ll \Gamma \) [6, 7], the resonance depths are modulated as \( S_1 \rightarrow S_1 \cos(\omega_m t + \phi) \). Feeding the modulated signal, along with the modulation, into a lock-in amplifier, the output in-phase component becomes

\[ V[\Delta] \propto \alpha_1 L_1 + \alpha_2 L_2 + O((G_S)^3), \]

(4)

where we evaluate the absorption coefficients on resonance:

\[ \alpha_1 = e^{-G_1(0 \rightarrow 2)} G_1(0 S_1), \]

and

\[ \alpha_2 = e^{-G_1(0 \rightarrow 1)} G_1(0 S_2). \]

Equation (4) describes two Lorentzians on a flat background, separated by the IS, with third-order nonlinear corrections to the small peak amplitudes. We now add phase modulation to the laser beam with a frequency much higher than the lock-in frequency \( \omega_M \gg \omega_m \). This creates sidebands in the pump and probe beams so that the resulting lock-in signal becomes as follows:

\[ V[\Delta] \propto \sum_{a=-\infty}^{\infty} \sum_{a'=-\infty}^{\infty} I_a J_a(\alpha_{ab} L_{ab} + \alpha_{ab} L_{ab}), \]

(5)

with \( I_a = I_a[m] \) as the Bessel function of order \( a \) with modulation index \( m \), and the sideband Lorentzians are \( L_{ab}[\Delta] = L_{ab}[\Delta - \omega_{M}]. \) Since they are independent of the laser frequency, we do not explicitly write the expressions for the peak amplitudes \( \alpha_{ab} \). The second term in (5) represents crossover peaks for each isotope obtained when the atoms are pumped by one sideband and probed by another [13]. There are no crossover peaks between different isotopes. Figure 2 shows the measured atomic signal presented in (5). We note that crossovers either fall between, or directly add to, the original peaks.

3. Frequency calibration method

In principle, it is possible to perform a wide scan similar to that presented in figure 2(a) and use the sideband peaks as markers for calibration of the frequency axis [14]; however, a wide scan is more prone to frequency drifts and relies on either a completely linear scan or a complete determination of the nonlinearity [8]. Instead, we scan only a small fraction of the actual separation, and calibrate the frequency axis using another modulated beam. When scanning the laser close to the second isotope resonance \( \Delta \approx \omega_{IS} \), and for a modulation frequency close to the IS, \( \omega_{M} \approx \omega_{IS} \) (region of interest in figure 2), only two peaks survive, which are separated by the difference between the modulation frequency and the IS

\[ V[\Delta] \propto \sum_{a=-\infty}^{\infty} \sum_{a'=-\infty}^{\infty} I_a J_a(\alpha_{ab} L_{ab} + \alpha_{ab} L_{ab}), \]

(6)

To have the remaining peaks at a similar size, we choose an appropriate modulation index \( m \approx 1 \). Generally, for a small
peak with amplitude \( a_2 \), and a larger one with \( a_1 \), and since the modulation index can be arbitrarily small, one can always choose \( m \) such that \((J_m/J_0)^2 \approx a_2/a_1\).

To accurately calibrate the frequency axis we split another beam, modulate its phase by \( \omega_{m} \), and insert it into an a Fabry-Pérot (FP) interferometer with finesse \( F \) and FSR \( \omega_{FSR} \). The transmitted intensity can be written as [15]

\[
I[\Delta] = I_0 \sum_{a=-\infty}^{\infty} a^2 I_1^a[\Delta, \omega_{FSR}/F],
\]

after filtering out terms oscillating at \( a\omega_{m} \) for \( a \neq 0 \). Equation (7) describes a series of Lorentzians, one for each sideband, separated by the modulation frequency. Figure 3 shows the lock-in signal in the region of interest, fitted with (6), along with the frequency calibration signal, fitted with (7).

4. Implementation

We use a narrowband (<MHz) single-frequency laser beam, from a home-made external cavity diode laser [16]. Frequency scanning is performed by applying voltage to a piezoelectric element connected to the external cavity grating. The beam is split in two. One part goes through a broadband, low-frequency (DC-100 MHz) commercial electro-optic modulator (EOM, New Focus 4002) and into an FP cavity (Thorlabs SA-200, \( \omega_{FSR} = 1.5 \text{ GHz} \), \( F = 0.0020 \)). The other part goes through a home-made, narrowband, high-frequency EOM [17], and enters a collinear, linearly polarized, pump-probe type setup with high-purity, natural abundance neon gas (90% \(^{20}\text{Ne} \), 9% \(^{22}\text{Ne} \), and 0.3% \(^{21}\text{Ne} \)), contained in an anti-reflection (AR)-coated glass cell residing in a high-Q coaxial resonator [18]. An RF-driven discharge at the resonance frequency (70 MHz) excites the atoms and populates higher-lying states. After ignition, a few milliwatts of RF power are sufficient to maintain stable plasma. The pump beam is amplitude-modulated by a chopper at \( \omega_c = 4 \text{ kHz} \). A reference beam goes through the cell as well, and provides another stage of subtraction to remove amplitude noise resulting from the laser (in part due to pointing instability and birefringent effects in the EOM) and cell discharge. The signal is fed into a lock-in amplifier (SRS SR830) where it is mixed with the chopper reference, filtered, and amplified. Figure 4 shows the main elements of the experimental system.

A slow (few Hz) and narrow (200 MHz) scan of the laser frequency results in simultaneous traces of the lock-in and FP signals (figures 2 and 3). We tune the relative FP frequency position by applying DC voltage to a piezoelectric element moving one of the cavity mirrors. From (6), the distance between the zero-order \(^{22}\text{Ne} \) peak and the first-order \(^{20}\text{Ne} \) is exactly \( \omega_d = \omega_M - \omega_{IS} \). We tune the low-frequency EOM to \( \omega_m \approx \omega_d \) by placing two of the FP sideband peaks directly on top of the lock-in atomic signal peaks (see figure 3). This limits the effects of scan nonlinearity in calibration of the frequency axis to less than a few kHz per trace. To each trace we fit the atomic signal with two Lorentzians of (6), and the FP signal with five Lorentzians corresponding to the 0, \( \pm 1, \pm 2 \) sideband orders observed (7). To account for non-homogeneous broadening, and thus accurately model the tails of the peaks, each Lorentzian in the fits is replaced with a pseudo-Voigt profile [19]. The fitting procedure gives the distances between the FP peaks \( \tau_{FP} \) and the atomic peaks \( \gamma_1 \) in units of time, and therefore the IS is calculated as follows:

\[
\omega_{IS} = \omega_M - \omega_d = \omega_M - \omega_m \frac{\gamma_1}{\tau_{FP}} \tag{8}
\]
This procedure of obtaining the IS is robust against frequency drifts in the laser (few MHz per minute), since both the atomic and FP signals drift together. The FP FSR is not used, and thus slow (few MHz per several minutes) thermal drifts in the cavity length serve only to move the FP signal relative to the atomic signal.

5. Results and discussion

For each experimental run, about 20 traces are taken with identical parameters (laser power, pressure, etc). The results are calculated using (8), and averaged using a Bayesian analysis approach with the Just Another Gibbs Sampler (JAGS) program [20, 21], which takes into account possible correlations between measurement errors and their intrinsic scatter.

To account for unknown systematic effects we investigate the calculated IS for different experimental parameters. By varying the laser power (figure 5(c)), we change the width and height of the peaks through saturation broadening [11]. By varying the RF discharge power (figure 5(b)), we change the excited-state population and peak height, as well as shifts that may result from non-thermal distribution of the gas sample. Hysteretic effects were observed at high RF power, where coupling of radio waves to the plasma changed from capacitive to inductive [22], and therefore we limited our investigation to low powers. The most stringent test for our measurement scheme is to vary both EOM frequencies together (figure 5(a)). This changes both the distance and magnitude of all peaks involved. The above measurements were obtained in a sealed cell at a pressure of 200 mTorr. We then replaced it with a glass tube with a gas inlet. The tube was first pumped to under 1 mTorr and then filled with high-purity, natural abundance neon gas at various pressures. The pressure drifted less than 1% during experimental runs. The results of this set of experiments are presented in figure 6. Although similar lines for $^{20}$Ne are expected to shift by about 2 MHz/Torr [23], no pressure shift in the IS was observed within our measurement uncertainties, which indicated that the shift is similar between the isotopes to a few 10 kHz per Torr.

The results of the sets presented in figures 5 and 6 are combined using the JAGS framework to obtain a weighted result of $1626.289 \pm 53$ MHz, where the quoted uncertainty range is one standard deviation.

We now discuss the contributions of some known systematic corrections, which are not affected by the parameters scanned, to the obtained experimental value. We note here that in our measurement scheme, the $^{22}$Ne peak appears at a lower laser frequency than the $^{20}$Ne peak (See figure 2). Due to their similar electronic configuration and identical quantum numbers, most of the systematic shifts between the lines of $^{20}$Ne and $^{22}$Ne vanish to high orders when measuring the IS. Among those are Zeeman shifts. The $^{3}P_{2}$ and $^{3}D_{3}$ levels in Ne are 24 and 5 THz away from their closest neighbors, respectively. Since the quantum interference shift is inversely proportional to the difference between levels [30], this effect is vanishingly small in our case. Naturally, the main difference between the two isotopes is their mass $M$. It affects atomic recoil, thus creating the so-called recoil shift of $\omega_{r} = \hbar/(2M\lambda^{2})$, a $-2.2$ kHz shift to the IS. The thermal distribution cancels first-order Doppler shifts but adds a second-order shift of $-4T/(\lambda c \pi M)$ [11], a negligible 75 Hz correction. The corrected result for the IS is thus $1626.287 \pm 53$ MHz.
6. Conclusion

We presented a simple measurement scheme for accurate determination of intervals between far-lying (up to few GHz) resonances in a spectroscopy signal. This method was used to measure the IS between the $^{20}\text{Ne}$ and $^{22}\text{Ne}$ cooling transition with high precision. Figure 7 and table 1 shows a comparison between the results presented here, and those of other groups using various experimental techniques. We note that earlier attempts [25–29], obtained a 4 MHz larger shift than those reported in this work and in [24]. It would thus be beneficial to conduct a high accuracy, ab initio calculation of this quantity, which as far as we know, does not exist in the literature [24].

To check our results with a different experimental system, we intend to conduct this measurement in our trap setup [31]. By measuring $^{21}\text{Ne}$ as well, it is also possible to improve determination of the $^{20–22}\text{Ne}$ charge radii difference [32].

Acknowledgments

This work was supported by the Israeli Science Foundation under ISF Grant No. (139/15); BO is supported by the Hoffman leadership and responsibility program, and the Eshkol Fellowship of the Ministry of Science and Technology.

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Table 1. IS and standard error obtained by various experimental techniques.

| Reference | Reported Value (MHz) | Method |
|-----------|----------------------|--------|
| This Work | 1626.287 ± 0.053     | Dual-sideband saturated absorption |
| [24]      | 1625.9 ± 0.15        | Trap absorption |
| [24]      | 1626.0 ± 0.22        | Trap fluorescence |
| [25]      | 1630 ± 3             | Velocity selective optical pumping |
| [26]      | 1631.2 ± 5.0         | Optogalvanic spectroscopy |
| [27]      | 1629.5 ± 1.0         | Intermodulated optogalvanic spectroscopy |
| [28]      | 1628 ± 3             | Doppler-free two-photon spectroscopy |
| [29]      | 1632 ± 3             | Supersonic beam |

Figure 7. IS and 68% confidence intervals obtained by different groups: (a) This work; (b) Feldcer et al [24], 1—Absorption, 2—Fluorescence; (c) Julien et al [25]; (d) Guthrohrlein et al [26]; (e) Basar et al [27]; (f) Odintsov et al [28]; and (g) Konz et al [29].