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Measurement of ionization in direct frequency comb spectroscopy

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Direct frequency comb spectroscopy is currently one of the most precise techniques for studying the internal structure of atomic and molecular systems. In this technique, a train of ultrafast laser pulses excites states in the target system which then relax, emitting fluorescence. The measured fluorescence is then plotted as a function of the comb parameters. But according to recent theory, the ultrashort pulses from the comb laser can also significantly ionize the target. Here, we test this theory by measuring the ion signal from direct frequency comb spectroscopy. Furthermore, instead of actively controlling the frequency comb parameters, we allow them to drift passively, measuring them and the ion signal simultaneously. The experiments were found to be in satisfactory agreement with theory, and the passive comb approach was found to be functional, though not as convenient as the conventional actively locked comb. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4794813]

I. INTRODUCTION

Laser excitation sets the standard for precision determination of atomic and molecular energy levels. With continuous wave (CW) lasers, the method is limited because of the need to relate the optical frequency of a measured interval to known secondary standards or to the primary rf standard of frequency. Roughly a decade ago, the field of optical laser spectroscopy was revolutionized by the realization of the optical frequency comb, which uses a mode-locked laser to produce a sequence of ultra-short light pulses, whose Fourier spectrum is a stable comb of discrete frequencies which are multiples of an rf frequency that can be directly compared to the primary frequency reference.[1,2]

In direct frequency-comb spectroscopy (DFCS), the pulsed laser light directly illuminates the target and the comb frequencies are scanned until one of the teeth excites the transition of interest. The excitation is generally detected by collecting the photons from spontaneous decay[3] of the excited state or by ionizing the excited state with a separate laser.[4] Because of the high intensity and broad bandwidth of the comb laser pulses, it is possible that the exciting laser could also ionize the excited state if it is energetically near enough to the continuum. Target ionization is the effect that is investigated in this work.

The basic idea of comb spectroscopy is that the frequency of the n^{th} tooth of the comb is given by

$$f_n = nf_{\text{rep}} - f_{\text{off}}, \tag{1}$$

where f_{rep} is the laser repetition frequency and f_{off} is the offset frequency. Then, when the combination of f_{rep} and f_{off} causes a comb tooth to be resonant with a transition between the ground state and a state in an excited manifold, the excited state is efficiently excited. Remarkably, it has also been shown[3,5,6] that states in higher manifolds can also be excited through non-resonant two-comb-tooth excitation, where by "non-resonant" we mean that while the sum frequency of the two comb teeth is resonant, the individual comb teeth are not. Therefore, by holding f_{off} fixed, and scanning f_{rep} one obtains a spectrum of either the first excited manifold of states or of the higher manifold, depending on the color of the fluorescence one detects. Those experimental results have been placed on firm theoretical ground.[7,8]

Considering that a frequency comb is a train of femtosecond pulses having high peak intensity, it is reasonable to ask if, in addition to the excitation, do frequency combs also significantly ionize the system. While this process has recently been investigated theoretically,[9,10] to our knowledge, it has not been investigated experimentally. The relevant questions are: "Is the ionization signal strong enough to use for DFCS?," and "If so, what information does an ionization spectrum contain?." These questions are addressed experimentally in the present work.

Ion detection has some advantages over photon detection. Application of a weak electric field enables 4π steradians collection angle for ions, whereas an experiment's geometry can greatly reduce the photon collection angle. Furthermore, ion detectors are generally more efficient and have lower dark count rates than photon detectors. In addition, photon detection can be somewhat problematic when the frequency of the emitted light is within the bandwidth of the comb laser; in this case, some care must be taken to reduce the background counts from scattered laser light. This is not an issue for ion detection.

In a recently published theoretical study of frequency comb light interacting with ^{87}Rb[10] Lomsadze *et al.* computed the relative populations of the 5*s*, 5*p*, 5*d*, and the continuum states. It was found that the structure of the ion spectrum was essentially identical to that of the 5*d*, and that the ion population was substantially greater. Figure 1 shows the relative populations of the 5*d* and continuum from those calculations. From Fig. 1, we can see that theory predicts the ion population to be about an order of magnitude greater than the 5*d* population. Furthermore, the structures in the ion spectrum are identical to those in the 5*d* spectrum, even over

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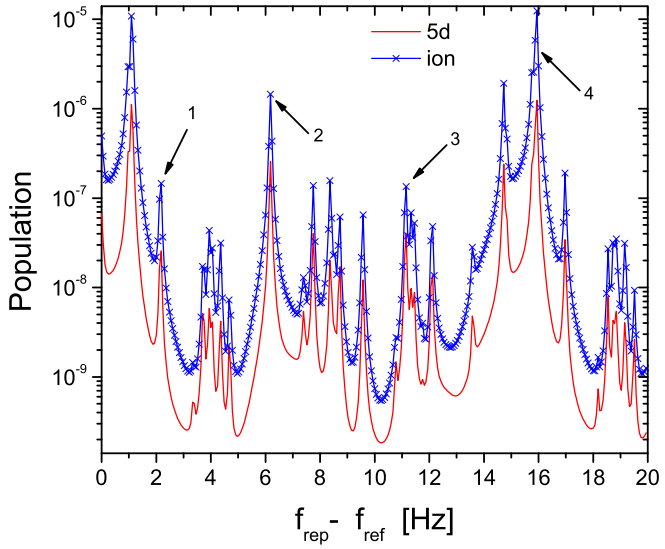

FIG. 1. Plot of the relative populations in the $5d$ and Rb^+ manifolds as functions of the comb laser repetition frequency, minus a fixed reference frequency, $f_{ref} = 75557551$ Hz, and $f_{off} = 14.5$ MHz. The $5d$ population is plotted as a solid line without points (red). The Rb^+ population is plotted as crosses connected by a line (blue). All of the peaks in the $5d$ and ion curves have been identified as resulting from 2-photon transitions from the $5s \rightarrow 5d$ manifolds. A few selected peaks are labeled as (1) $5s_{1/2}, F = 1 \rightarrow 5d_{5/2}, F = 2$; (2) $5s_{1/2}, F = 2 \rightarrow 5d_{5/2}, F = 2$; (3) $5s_{1/2}, F = 1 \rightarrow 5d_{5/2}, F = 1$; (4) $5s_{1/2}, F = 2 \rightarrow 5d_{5/2}, F = 4$.

the several orders of magnitude dynamic range showed in the figure. Figure 1 suggests that one can do DFCS by measuring ions rather than photons.

The goal of the present work is two-fold. First, we wish to measure the ionization signal as a function of the laser repetition rate and offset frequency. This will address the two fundamental questions mentioned above. At the same time, we test a novel methodology in which the experiment is run with a free-running oscillator, rather than with a tightly controlled frequency comb.

II. EXPERIMENTAL SETUP

The experimental setup is a slight modification to our MOTRIMS[11] (magneto-optical trap recoil ion momentum spectroscopy) apparatus. The light from a Ti:sapphire oscillator, running at a nominal repetition rate of 80 MHz, is directed onto a target of ^{87}Rb that is trapped and cooled to about 200 μK using a magneto optical trap (MOT).[12] The sole purpose in cooling the atoms is so that Doppler effects can be neglected. In the normal course of cooling and trapping, an average of approximately 20% of the trapped atoms were in the $5p_{3/2}, F = 3$ state[13] with most of the remaining atoms in the $5s_{1/2}, F = 2$ state. Because the principle goal of this experiment was to see if the ion signal was appreciable we were not overly concerned with resolution. We, therefore, did not turn off the trapping magnetic field gradients. The resulting broadening due to the Zeeman effect is estimated to be 2 MHz which corresponds to 0.39 Hz on a DFCS spectrum. Furthermore, we did not worry about power-broadening by the comb laser which is estimated to be 5 MHz (0.98 Hz on a DFCS spectrum). The trapping lasers were left on because we felt the resulting distortions to the DFCS spectra were not relevant to our goals and leaving them on significantly improved our measurement duty cycle.

The oscillator is free running, meaning that no controls are placed on either f_{rep} or f_{off}. A rapidly varying f_{rep} and/or f_{off} would destroy the essential properties of a comb, namely the regularly spaced, nearly delta-function structures in the frequency domain. However, a direct measurement of the temporal evolution of f_{off} using the usual $f - 2f$ interferometer,[1,2] and of f_{rep}, both measurements made with precision counters[14] that were referenced to a GPS signal, showed that drift in those parameters was slow. Over a 1-h time interval, the typical net drift in f_{rep} and f_{off} were 30 Hz and 2 MHz, respectively. From Eq. (1), this drift gives rise to a range Δf_n of about 150 MHz, for $n \sim 5.1 \times 10^6$. This is more than adequate to cover the desired tuning range for spectroscopic studies of rubidium.

In Fig. 2, we plot the Allan deviations in f_{rep} and f_{off} as functions of the sampling time. We see that in a 1 s sampling time, the maximum deviation in f_{rep} was under 0.3 Hz and for f_{off} the deviation was under 300 kHz. This gives an uncertainty in comb tooth frequency of less than 1.7 MHz. Furthermore, if we extrapolate the Allan deviation in f_{rep} down to 100 ms, the shortest period over which our counters can sample with adequate precision, we would expect an Allan deviation of about 0.2 Hz, leading to a comb tooth uncertainty of about 1 MHz. This is not much broader than the 0.7 MHz line width[15] of the states in the $5d$ manifold of Rb. The basic idea, then, was a passive approach to DFCS in which, due to thermal effects, the comb slowly auto-scanned both f_{rep} and f_{off}. These parameters were measured every 100 ms and were recorded along with the ion signal.

Rubidium ions, produced through photoionization of excited states by the comb laser light, were extracted from the MOT region by the application of a weak ($\sim$10 V/cm) electric field. The ions were detected by a micro-channel plate (MCP) with resistive anode. The detector's efficiency is nearly 50%, limited by the hole-to-open area ratio of the MCP. Because of the electric field all the ions, regardless of emission angle, are directed to the detector. The geometry of

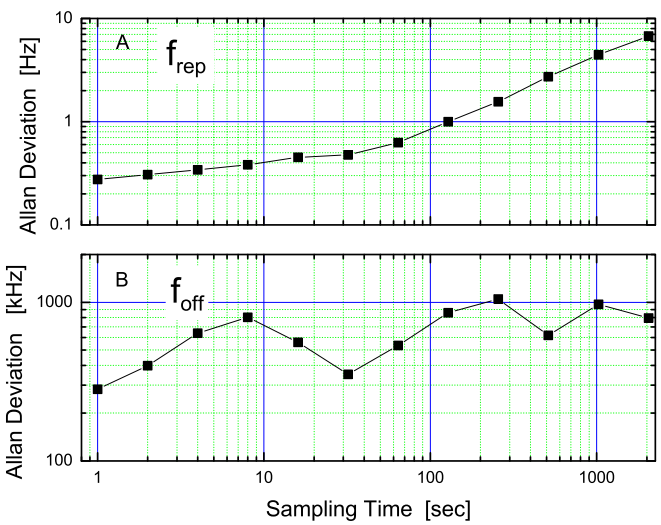

FIG. 2. The Allan deviations of f_{rep} (A) and f_{off} (B) are plotted versus sampling time.

the MOTRIMS apparatus makes photon detection virtually impossible.[16] Therefore, a direct quantitative comparison of ion and photon collection rates was not possible, but the ion count rate is clearly significant.

III. RESULTS

Figure 3 shows a typical experimental result. This represents 130 min of data collection with an average laser power of 25 mW focused to beam diameter of 5.7×10^{-4} m. The laser pulse width was 50 fs. The density in the figure represents the average ion count rate at each set of parameters f_{rep} and f_{off}. Since the laser was allowed to drift passively, f_{rep} and f_{off} do not necessarily fill out the full phase space represented in the plot, i.e., a zero count rate could mean that no ions were generated when the laser was in that range, or it could mean that the laser never drifted into the range represented. The data from the experiment are made up of a stream three-value vectors each accounting for 100 ms of elapsed time. The vectors are composed of an f_{rep} measurement, an f_{off} measurement, and the number of ion counts accumulated during the measurement period. The data are then run through a routine that divides the parameter space into bins of width 0.1 Hz in f_{rep} and 2 MHz in f_{off} and sums the ion counts into the appropriate bin as each datum is read. After binning, the ion count total in each bin is divided by the number of data points that were summed into that bin in order to get the ion production rate. It is these ion production rates that are plotted in Fig. 3.

Ionization in Fig. 3 appears as nearly vertical stripes. This is because of the relationship between f_{rep} and f_{off} expressed by Eq. (1). For a single-photon resonance at frequency f_t, all f_{rep} and f_{off} satisfying $nf_{rep} - f_{off} = f_t$ will produce enhanced ionization. Thus, resonances appear as lines with slope n in Fig. 3. For our laser, n is about 5.1×10^6, so a change of 1 MHz in f_{off} would shift a resonance peak 0.2 Hz in f_{rep}. For two-photon resonances, the stripes will have slopes given by the average of the tooth orders of the two transition frequencies. In principle, we could determine n by fitting the two-dimensional data in Fig. 3. This would not yield very good precision in our case both because of our experimental resolution and because of the random nature of our parameter space coverage.

We choose, for demonstration purposes, to condense the data to one dimension by projecting to the f_{rep} axis along diagonals of slope n, where n is determined from published data. We further arbitrarily choose to project to a value of $f_{off} = -23.5$ MHz. A further compression of the data can be made by noticing that Eq. (1) is cyclic in the order n. Thus, if two repetition frequencies are related by $nf_1 = (n+1)f_2$, both could excite the same atomic resonance, for constant f_{off}. For our laser, a given one-photon transition should repeat at intervals of approximately 14.9 Hz in f_{rep}. We can, thus, combine repeated lines in our projected spectrum into a single range by cutting the f_{rep} axis at intervals of 14.9 Hz and shifting segments to the left by multiples of the cyclic interval. Each ion rate is averaged over the number of segments combined this way. The projected data are shown in Fig. 4.

The solid curve in Fig. 4 is from the measured ion rates, while the crosses are theoretical calculations scaled to match the measured data. The theory is the same as used for Fig. 1, but with the laser power set to the experimental value and f_{rep} and f_{off} also matched to the range of the measurements. We note that adding cycles in f_{rep} and f_{off} can cause the relative heights of the peak to change due to the fact that different cycles corresponding to different f_{rep} and f_{off}, have different detunings from the intermediate states. While this variation in peak heights makes comparison with theory more difficult (because the spectrum was calculated with a single value of f_{off}) the *positions* of the structures in the spectra are unaffected by this averaging. The projection routine is not exact and contributes an additional width of approximately 0.1 Hz to the peaks in the DFCS scan, which is minor compared to the experimental resolution of 1 Hz. We note

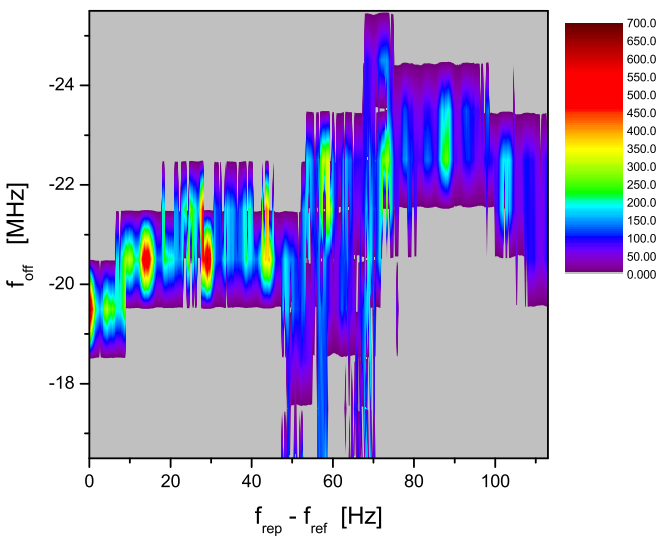

FIG. 3. Plot of rubidium ion count rate, in ions per 100 ms, as a function of the freely varying parameters f_{rep} and f_{off}. $f_{ref} = 75616231$ Hz.

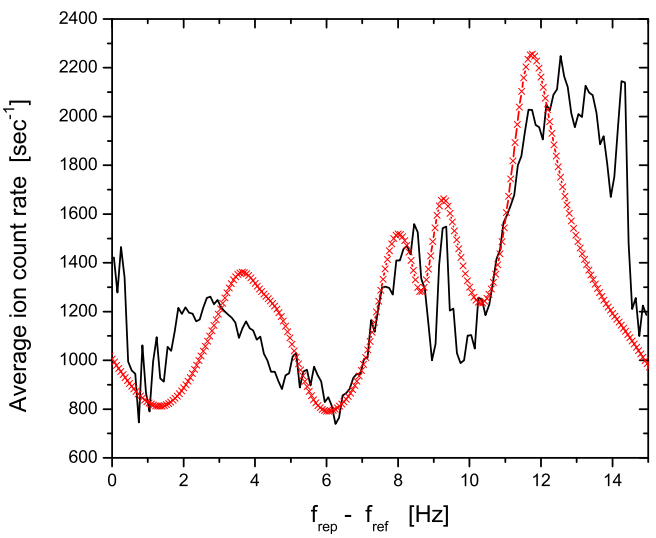

FIG. 4. The data from Fig. 3 (solid black line). The ion count rates have been averaged over the cycles in f_{rep} and f_{off} and plotted versus $f_{rep} - f_{ref}$, where $f_{ref} = 75616232$ Hz. Also plotted is the calculated ion signal rate (crosses connected by a line, red). For both theory and experiment $f_{off} = -23.5$ MHz.

that this kind of analysis is not needed in active DFCS since there is no reason to simultaneously scan both f_{rep} and f_{off}, nor is it necessary to scan f_{rep} over multiple orders.

One cannot help but notice that the data of Fig. 4 are considerably broader than the calculations shown in Fig. 1. We emphasize that the power broadening was a result of our conveniently using a power level that resulted in usable counting statistics over a time that we could keep the f_{off} measurement stable. It is not a limitation of the ion-detection technique, which could be used with active DFCS, as well. The information content achieved with ion counting would be identical with that achieved with photon counting, under similar laser intensity conditions.

From Fig. 4, we can see that we obtain a large ionization rate, clearly adequate for spectroscopic measurements. There is also clear structure in the graph that is consistent with the calculated ionization spectrum. The differences between theory and experiment are due to the trapping laser being left on. For example, the apparent shift of some of the experimental peaks toward lower frequencies (near 2 Hz) and higher frequencies (near 13 Hz) are actually due to enhanced excitation of the $5p_{3/2}$, $F=3$, by the trapping laser. Photoionization of the 5*d* states by the trapping laser is also possible. We believe this is negligible because the photoionization rate is proportional to laser intensity and the trapping laser's average intensity is more than 3 orders of magnitude less than that of the comb laser.

IV. SUMMARY

In this work, we constructed and ran an experiment that showed that ionization is an important process in DFCS. Furthermore, we demonstrated that the experimental and theoretical ionization spectra are consistent. Furthermore, from earlier work and from Fig. 1, we know that the theoretical ionization spectra are essentially identical to the excitation spectra. This was not *a priori* obvious because one could imagine a scenario in which one comb tooth excites a state in the 5*p* manifold, and two other teeth combine to "resonantly" photoionize that 5*p* state. We note that for this work atomic Rb was used as a test system. But the technique can be used to investigate any state of any system of interest so long as that state can be accessed using two photons within the comb laser bandwidth, and that state can be photoionized by one additional photon from the comb laser, in analogy with the cases shown here for Rb.

Our goal was to demonstrate the strength of the ionization channel rather than the resolution for which frequency comb spectroscopy is already known. By following, the standard procedure of taking a series of measurements with gradually lower comb laser power and then extrapolating those spectra to zero intensity we would eliminate power broadening effects.

While our data were in satisfactory agreement with theory, there were some discrepancies. These were largely caused by processes due to the trapping lasers and were therefore irrelevant to the hypothesis we were testing. Just as in the case of the B-field gradient, the trapping laser could have been chopped to eliminate this effect, though the starting point in the comb laser excitation would still have been with optically pumped rubidium in the ground state. But the loss in duty cycle would have given rise to reduced counting statistics. Other slight distortions in the experimental spectrum were the result of averaging spectra over the range of f_{rep} and f_{off} (in going from Fig. 3 to Fig. 4) due to the different cycles having different detunings from intermediate *p* states. This gives rise to a near randomization in the relative peak heights, though their locations should remain unaffected.

Finally, we have demonstrated a novel implementation of DFCS in which f_{rep} and f_{off} were not controlled but varied through the natural interaction of the laser system with the changing ambient temperature. Our assessment of this passive comb is that, while workable, it has significant disadvantages over a conventional comb. The most serious of these is that one cannot simply dial in a tooth frequency of interest. Thus, we could not look for double resonances, in which one comb tooth was resonant with a transition between the 5*s* and 5*p* manifolds, while another tooth was resonant with a transition between the 5*p* and 5*d* manifolds. This makes it virtually impossible to use the passive comb to investigate, for example, optical pumping effects. Nevertheless, for laboratories that have an ultrafast oscillator but no true comb, impromptu passive comb experiments could still be done.

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