Absolute frequency measurement of the $6s6p\,^1P_1$–$6s7s\,^1S_0$ transition of ^{174}Yb in a Yb hollow-cathode lamp

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We report absolute frequency measurement of the $6s6p\,^1P_1$–$6s7s\,^1S_0$ transition at 1077.3 nm of ^{174}Yb in a hollow-cathode lamp (HCL). The frequency of coupling laser was stabilized to the $6s^2\,^1S_0$–$6s6p\,^1P_1$ transition at 398.9 nm using an Yb atomic beam, and the coupling laser beam was then overlapped with the counterpropagating probe laser beam at 1077.3 nm in the Yb HCL to obtain a Doppler-free signal of laser-induced birefringence for the frequency stabilization of the probe laser. Systematic frequency shifts of both transitions as a function of the discharge current of the Yb HCL were analyzed using an Yb-doped fiber laser frequency comb. The absolute frequency of the $6s6p\,^1P_1$–$6s7s\,^1S_0$ transition was measured to be 278 281 521.4(7.2) MHz, and from the published frequency of the transition, the absolute frequency of the $6s^2\,^1S_0$–$6s7s\,^1S_0$ two-photon transition at 291.1 nm was determined to be 1 029 807 509.2(7.2) MHz with the relative standard uncertainty of 7.0×10^{-9}.

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I. INTRODUCTION

Since the first demonstration a decade ago, optical frequency combs have played a major role in not only measuring absolute optical frequencies and fundamental physical constants [1–3], but also developing optical lattice clocks and single ion clocks [4,5]. In addition to recent efforts to extend the spectrum into unexplored regions such as the extreme ultraviolet [6], there is an increasing interest in investigating the Rydberg transitions of various atomic systems [7] for applications of quantum information technology based on Rydberg blockade [8]. Furthermore, narrow stepwise excited-state transitions of alkaline-earth-metal atoms have been proposed for second-stage cooling to achieve micro-Kelvin temperatures [9]. Precise measurements of the transition frequencies between excited atomic states are therefore essential for such applications [7] as well as for experimental tests of the theoretical values determined using different calculation methods for the excited state energies of rare-earth atoms [10,11].

The cascade two-photon $6s^2\,^1S_0$–$6s6p\,^1P_1$–$6s7s\,^1S_0$ transition of Yb has been investigated extensively for two-photon laser cooling [9] and efficient generation of a pair of correlated two photons [12,13]. Here we report on our efforts to measure the cascade two-photon transition frequencies of ^{174}Yb in an Yb hollow-cathode lamp (HCL). To measure the absolute frequency of the upper probe transition, we first stabilized the frequency of a coupling laser to the $^1S_0$–$^1P_1$ transition by using an Yb atomic beam, for which a Doppler-free resonance fluorescence signal was used for the frequency stabilization. The coupling laser beam was then overlapped with the counterpropagating probe laser beam through the hollow channel of the Yb HCL. A modulation-free dispersive signal, induced by the circular birefringence of the coupling laser in the Yb HCL, was used to stabilize the frequency of the probe laser [13]. Finally, systematic frequency shifts depending on the discharge current of the Yb HCL were carefully investigated by using an Yb-doped fiber laser frequency comb (YLFC).

As a result, we were able to measure the absolute frequency of the $6s6p\,^1P_1$–$6s7s\,^1S_0$ transition to be 278 281 521.4(7.2) MHz. In addition, from the reported value of the $^1S_0$–$^1P_1$ transition frequency at 398.9 nm [14], we determined the absolute frequency of the $6s^2\,^1S_0$–$6s7s\,^1S_0$ two-photon transition at 291.1 nm to be 1 029 807 509.2(7.2) MHz with a relative standard uncertainty of 7.0×10^{-9}. The experimental results reported here for the excited state energies of ^{174}Yb agree well with the most accurate theoretical values calculated using the Hartree-Fock relativistic (HFR) method including the valence-valence correlation [11].

II. FREQUENCY STABILIZATION OF COUPLING AND PROBE LASERS

The atomic energy levels relevant to the current experiment are shown in Fig. 1(a). The ground state ($|0\rangle$) of the ^{174}Yb atom is the $6s^2\,^1S_0$ state, which is coupled to the $6s6p\,^1P_1$ intermediate state ($|1\rangle$) through the coupling laser at 398.9 nm and frequency f_c. State $|1\rangle$ is also coupled to the $6s7s\,^1S_0$ state ($|2\rangle$) through the probe laser at 1077.3 nm and frequency f_p. The $|0\rangle$–$|1\rangle$ transition is an E1 transition with a broad natural linewidth of $\Gamma = 2\pi \times 28$ MHz and has been used for first-stage laser cooling and trapping of Yb atoms [15,16]. The upper $|1\rangle$–$|2\rangle$ transition is also an E1 transition but with a narrow natural linewidth of $\Gamma = 2\pi \times 3.5$ MHz [17] and has been proposed for two-photon laser cooling [9] and efficient generation of a pair of correlated two photons via the spin-triplet $6s6p\,^3P_1$ state [12]. The $|0\rangle$–$|2\rangle$ transition (291.1 nm) is a forbidden transition for the electric dipole.

Figure 1(b) shows the experimental setup for the frequency stabilization of the coupling and probe lasers as well as the absolute frequency measurements of both transitions using the YLFC developed in-house. A grating-stabilized extended-cavity diode laser (ECDL) at 398.9 nm was used as the coupling laser to drive the $|0\rangle$–$|1\rangle$ transition, while an ECDL at 1077.3 nm was used as the probe laser to drive the $|1\rangle$–$|2\rangle$ transition. In this experiment, an Yb atomic beam and Yb HCL were used for the frequency stabilization of the coupling and probe lasers, respectively, as described in previous works

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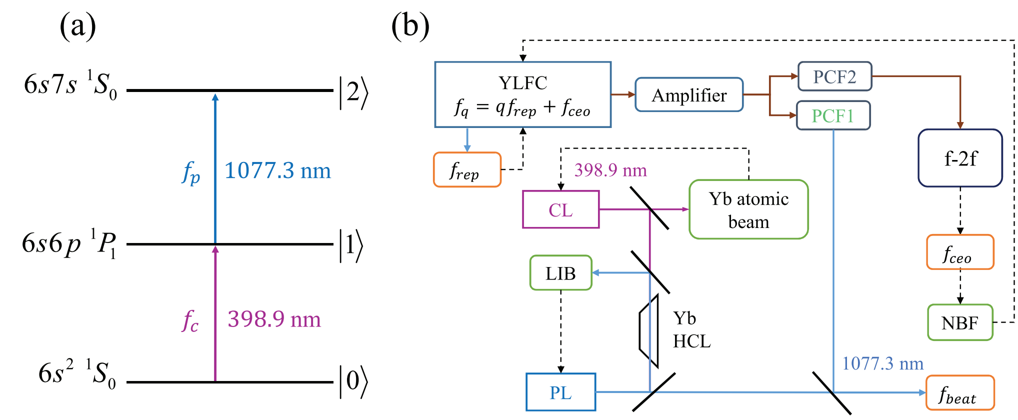

FIG. 1. (Color online) (a) Energy-level diagram of Yb showing the states involved in the cascade two-photon transition. (b) Experimental setup. CL: coupling laser at 398.9 nm of frequency f_c; PL: probe laser at 1077.3 nm of frequency f_p; PCF: photonic crystal fiber; $f-2f$: self-referenced $f-2f$ interferometer; NBF: narrow bandpass filter; LIB: modulation-free signal detector of laser-induced birefringence. Solid and dashed lines represent laser beams and electrical connections, respectively. Other symbols are described in the text.

[12,13]. Here we briefly describe the features of the two methods.

The frequency of the coupling laser was stabilized to the peak of the resonance fluorescence signal scattered from the intermediate state $|1\rangle$ of ^{174}Yb in the Yb atomic beam [12]. The linewidth of the measured resonance fluorescence signal was 54 MHz. Then, the frequency of the probe laser was stabilized through a Doppler- and modulation-free spectroscopy of laser-induced birefringence (LIB) in the Yb HCL [13]. A dispersive LIB signal with a linewidth of 90 MHz was measured. As the frequency of the coupling laser was locked to the center of the resonance fluorescence signal, it does not depend on the operation conditions of the Yb HCL. Within the Yb HCL, however, the LIB signal exhibited a slight asymmetry due to the pressure shifts of the excited $|1\rangle$ and $|2\rangle$ state energies, which depend on the operating current of the Yb HCL. Therefore, in the experiment, we adjusted the LIB signal so that it was symmetric for each operation current of the Yb HCL, which resulted in a major systematic frequency shift of the $|1\rangle$–$|2\rangle$ transition (see Sec. IV).

The frequency stabilities of the two lasers for the two-photon transition in the Yb HCL were measured to be ±300 kHz by analyzing the beat frequency stability between the probe laser and the nearest comb component of the YLFC.

III. FREQUENCY MEASUREMENT WITH YLFC

The absolute frequency of the $6s^2\ ^1S_0$–$6s6p\ ^1P_1$ transition of ^{174}Yb was measured previously using an effusive Yb atomic beam system [14], and a Rb-stabilized ring-cavity resonator was used to measure the absolute frequencies of various isotopic components in the 399-nm $^1S_0$–$^1P_1$ line of Yb. They reported a transition frequency value of $f_{\mathrm{CL}}^{D} = 751\,525\,987.761(60)$ MHz with a relative standard uncertainty of 8.0×10^{-11}. We used this published value for an indirect determination of the absolute value of the two-photon $6s^2\ ^1S_0$–$6s7s\ ^1S_0$ transition.

To determine the absolute frequency of the excited $|1\rangle$–$|2\rangle$ transition, we measured the beat frequency f_{beat} between the probe laser and the nearest frequency comb component of the YLFC, as shown in Fig. 2(b). The linear-cavity oscillator of the YLFC has a repetition frequency of $f_{\mathrm{rep}} \cong 188$ MHz (Fig. 2) and an output power of 73 mW [18]. The frequency f_q of the qth mode of the YLFC is given by $f_q = qf_{\mathrm{rep}} + f_{\mathrm{ceo}}$, where f_{ceo} is the carrier-envelop offset frequency of the YLFC [1,2]. Stabilization of f_{rep} was achieved by using a phase-error signal detected at the fifth harmonic frequency of $5f_{\mathrm{rep}} \cong 941$ MHz with reference to a frequency synthesizer [Fig. 3(a)]. More precisely, the phase-coherent link to the SI frequency standard was achieved by locking the time base of the frequency synthesizer to the 10-MHz reference signal of an oven-based Rb clock, whose time base was in turn locked to the clock signal of the global positioning system [19]. Frequency counters used in this experiment were also linked to the same SI time base.

A single-stage Yb-doped fiber amplifier [Fig. 1(b)] with a 10-μm-core diameter fiber (Liekki Yb1200-10/125DC-PM) was used to increase the output power of the oscillator up to 3 W. A pair of transmission grating (not shown) was used following the amplifier for frequency dechirping, before launching the output beam into two photonic crystal fibers (PCF1 and PCF2) for spectral broadening. The optical power and pulse width of the amplified and dechirped YLFC were measured to be 1.5 W and 180 fs, respectively. An output power of approximately 1 W was launched into PCF1 (NKT Photonics; NL-800-PM) for the $f-2f$ interferometer, while 0.8 W was launched into PCF2 (NKT Photonics; NL-1050-zero-2) for the f_{beat} measurement. The features of the spectral broadening over one octave with a flat-top spectrum are described elsewhere [19].

Figures 2(a) and 2(b) show the measured spectra of f_{ceo} of the FLFC and f_{beat}, respectively, within one free-spectral range of f_{rep}. The measured signal-to-noise (S/N) ratio and linewidth of f_{ceo} were 20 dB and 15 MHz, respectively. To stabilize and measure f_{ceo}, we used a narrow band-pass filter (NBF) with a 2-MHz bandwidth and a center frequency that could be tuned from 50 to 95 MHz. We modulated the pump power of the oscillator of the YLFC with a 150-kHz signal and obtained a frequency discriminator signal by using a high-frequency lock-in amplifier (Stanford Research Systems; SRS844). A typical dispersive signal suitable for the frequency

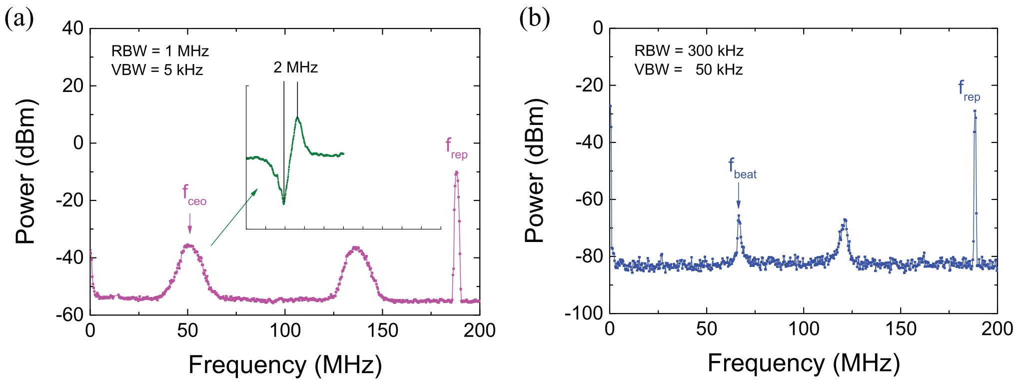

FIG. 2. (Color online) (a) Carrier-envelop offset frequency $f_{\rm ceo}$ of the YLFC measured using the $f-2f$ interferometer in Fig. 1(b) with a linewidth of 15 MHz. Inset: Frequency discriminator signal obtained with the NBF in Fig. 1(b). (b) Measured beat frequency $f_{\rm beat}$ between the probe laser at 1077.3 nm and the nearest comb component of the YLFC.

stabilization of $f_{\rm ceo}$ is shown in the inset of Fig. 2(a). The error signal was fed back into the injection current of the pump laser of the oscillator to stabilize $f_{\rm ceo}$. In this way we achieved an Allan deviation of $\sigma_y = 5\times10^{-3}$ (±250 kHz at a 1 s gate time), as shown in Fig. 3(d). The measured $f_{\rm beat}$ between the probe laser and the nearest comb component of the YLFC had an S/N ratio of more than 20 dB, as shown in Fig. 2(b).

After stabilizing both $f_{\rm rep}$ and $f_{\rm ceo}$ of the YLFC, $f_{\rm beat}$ was measured at various discharge voltages of the Yb HCL. Figures 3(a), 3(b), and 3(c) show the results of the frequency measurements for $f_{\rm rep}$, $f_{\rm beat}$, and $f_{\rm ceo}$, respectively, as a function of time. The Allan deviation of $f_{\rm rep}$ was $\sigma_y(\tau) = 1.2\times10^{-12}\tau^{-1/2}$, resulting in an optical frequency fluctuation of $\delta(qf_{\rm rep}) = \pm5.0$ kHz at a 1 s gate time, where $q = 1479\,230$ (see below). We set the current of the Yb HCL to $I_{\rm HCL} = 1.74$ mA ($V_{\rm op} = 163$ V) to measure $f_{\rm beat}$, and the Allan deviation of $f_{\rm beat}$ was $\sigma(\tau) = 5.7\times10^{-3}\tau^{-1/2}$, resulting in a statistical uncertainty of ±320 kHz at a 1 s gate time.

We obtained, from Fig. 3, the mean values of $f_{\rm rep}$, $f_{\rm beat}$, and $f_{\rm ceo}$ as well as the $1-\sigma$ sample standard deviations at the injection current of Yb HCL $I_{\rm HCL} = 1.74$ mA:

$$f_{\rm rep} = 188\,125\,856.9962(26)\ \text{Hz}, \tag{1a}$$

$$f_{\rm ceo} = 49.73(60)\ \text{MHz}, \tag{1b}$$

$$f_{\rm beat} = 56.36(30)\ \text{MHz}. \tag{1c}$$

Since $f_{\rm beat} = |f_{\rm PL} - f_q|$, where f_q is the frequency of the qth mode of the YLFC, the frequency $f_{\rm PL}$ of the probe laser can

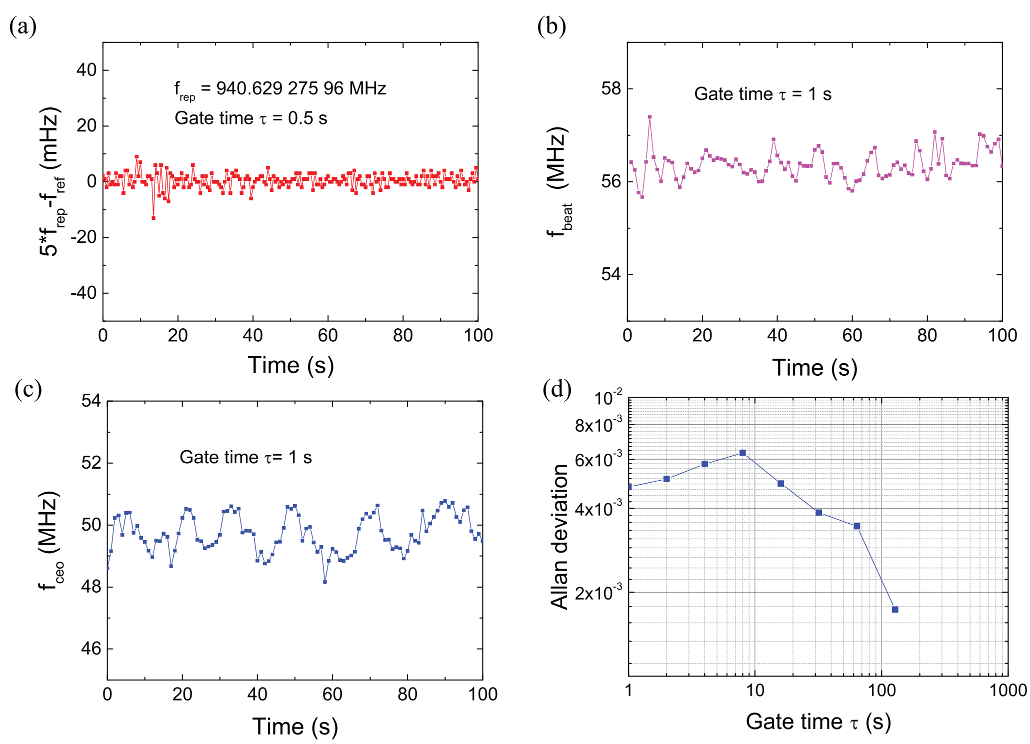

FIG. 3. (Color online) Measured (a) $f_{\rm rep}$, (b) $f_{\rm beat}$, and (c) $f_{\rm ceo}$ as a function of time. In (b), the operating current of the Yb HCL was 1.74 mA. (d) Allan deviation of $f_{\rm ceo}$ associate with the frequency fluctuation in (c).

be determined from the relation

$$f_{\rm beat} = |f_{\rm PL} \pm q f_{\rm rep} \pm f_{\rm ceo}|. \quad (2)$$

The signs in Eq. (2) and the absolute mode number q can be determined easily by changing $f_{\rm rep}$ by small amount and monitoring the change in $f_{\rm beat}$ [20]. We found that the mode number q corresponding to the measured values in Eq. (1) was $q = 1\,479\,230$.

IV. SYSTEMATIC EFFECTS AND FREQUENCY DETERMINATION

To study the systematic frequency shift of the probe transition at 1077.3 nm, we first measured the frequency shift of the intermediate state $|1\rangle$ as a function of the operating current of the Yb HCL. To do this, a Doppler-free polarization spectroscopy setup with an independent ECDL emitting at 398.9 nm was constructed [not shown in Fig. 1(b)] with the same Yb HCL (Hamamatau; L2783-70NE-Yb) used in Sec. III. The oscillation frequency f_s of the new ECDL was stabilized to the center of the polarization dispersion signal of the $|0\rangle$–$|1\rangle$ transition [21]. The beat frequency f_{sc} between $f_s = f_c'$ and f_c of the coupling laser in Fig. 1(b) was measured as a function of the driving current of the Yb HCL.

Figure 4(a) presents the results of the beat frequency measurements of f_{sc} of the coupling laser and $f_{\rm beat}$ of the probe laser. In Fig. 4(a) we see that f_{sc} and $f_{\rm beat}$ decrease as we increase the driving current of the Yb HCL. In particular, the amount of frequency shift of $f_{\rm beat}$ is observed to be five times larger than that of f_{sc} within the measurement current range, so that the major component of systematic uncertainty is caused by the extrapolation error of $f_{\rm beat}$ as described below. In addition, from these observations, we understand that the energies of level $|1\rangle$ and $|2\rangle$ in the Yb HCL are, respectively, shifted toward the lower values as we increase the current of the Yb HCL as described in Fig. 4(b).

The frequencies $f_{\rm rep}$, $f_{\rm ceo}$, and $f_{\rm beat}$ in Eq. (1) were measured at $I_{\rm HCL} = 1.74$ mA as explained in Sec. III. Therefore, to determine the absolute frequency of the $|1\rangle$–$|2\rangle$ transition, we need to extrapolate $f_{\rm beat}$ to the operation condition of $I_{\rm HCL} = 0$ because of the nonzero pressure shift when $I_{\rm HCL} \neq 0$. This was found to be the largest component among the various systematic effects of this study.

As described in Sec. II, $f_{\rm PL}$ was stabilized to the center frequency of the LIB signal obtained from the Yb HCL. Therefore, $f_{\rm beat}$ contains the frequency shift of not only the intermediate state $|1\rangle$ but also the upper state $|2\rangle$ at a nonzero driving current of the Yb HCL, as shown in Fig. 4(a). To estimate the value of $f_{\rm beat}$ at $I_{\rm HCL} = 0$, however, we need to know the asymptotic value $f_{\rm beat}^a$ because $f_{\rm beat}$, which is affected by both $|1\rangle$ and $|2\rangle$, converges to the asymptotic value. Finally, we estimated $f_{\rm beat}^a$ from the best fit with a single-exponential function, as shown in Fig. 4(a), to be

$$f_{\rm beat}^a = 60.1(7.2)\ \text{MHz}. \quad (3)$$

Here the systematic uncertainty of 7.2 MHz was obtained from the standard deviation of the mean of fitting error, i.e., $\sigma_s/\sqrt{n} = 7.2$ MHz, where $\sigma_s = 17.6$ MHz is the sample standard deviation and $n = 6$ is the number of data points in Fig. 4(b). Note that the systematic uncertainty of 7.2 MHz is one order of magnitude larger than the statistical uncertainties listed in Eq. (1).

We investigated other systematic effects such as Rabi frequencies of the driving lasers within the Yb HCL, but no noticeable frequency shift larger than the statistical uncertainties listed in Eq. (1) was observed.

V. RESULTS AND DISCUSSIONS

We used Eqs. (1) and (2) along with the asymptotic value of $f_{\rm beat}^a$ in Eq. (3), rather than $f_{\rm beat}$ in Eq. (1c) to determine the absolute frequency f_{12} of the $6s6p\,^1P_1$–$6s7s\,^1S_0$ transition. The combined uncertainty includes statistical and systematic uncertainties and was calculated to be $u_c = 7.2$ MHz, which is dominated by the systematic uncertainty of Eq. (3). We obtained an absolute value of the transition frequency of the

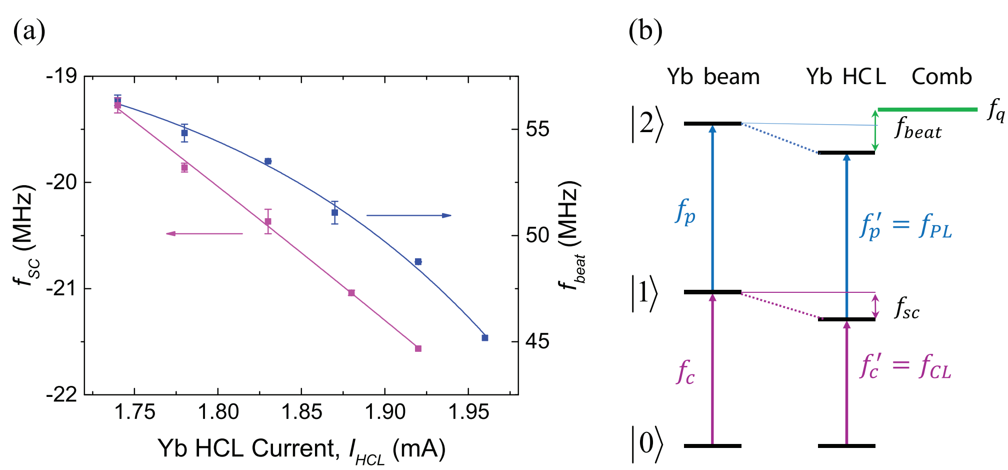

FIG. 4. (Color online) (a) Measured frequency shifts of the beat frequencies f_{sc} (circles) and $f_{\rm beat}$ (squares), respectively, as a function of driving current $I_{\rm HCL}$ of the Yb HCL. Error bars are sample standard deviations of each data set and solid lines are best fits of a single-exponential function. (b) Schematic illustration of the energy-level shifts of the intermediate $|1\rangle$ (f_c') and upper $|2\rangle$ (f_p') states in the Yb HCL compared to those of the same levels $|1\rangle$ (f_c) and $|2\rangle$ (f_p) in the Yb atomic beam, where f represents the transition frequency and f_q is the frequency of the qth mode of the YLFC.

probe laser of

$$f_{12} = 278\,281\,521.4(7.2)\ \text{MHz}, \tag{4}$$

with a relative standard uncertainty of $u_c/f_{\text{PL}} = 2.6 \times 10^{-8}$. The uncertainty is two orders of magnitude larger than the reported value of 8.0×10^{-11} associated with the absolute frequency measurement of the coupling laser in the Yb atomic beam [14]. Thus, future experiments with either an Yb atomic beam [12] or optically trapped Yb atoms [16] for the frequency stabilization of the probe laser are highly desirable to increase the measurement accuracy by reducing the uncertainty associated with the pressure shift in the Yb HCL.

By combining the absolute frequency of the $6s6p\,^1P_1$–$6s7s\,^1S_0$ transition in Eq. (4) with the published value of the $6s^2\,^1S_0$–$6s6p\,^1P_1$ transition of ^{174}Yb [14], we determined the absolute frequency f_{02} of the $6s^2\,^1S_0$–$6s7s\,^1S_0$ transition with a combined uncertainty of 7.2 MHz to be

$$f_{02} = 1\,029\,807\,509.2(7.2)\ \text{MHz}, \tag{5}$$

with a relative standard uncertainty of 7.0×10^{-9}.

The absolute frequency of Eq. (5) for the $6s^2\,^1S_0$–$6s7s\,^1S_0$ two-photon transition at 291.1 nm of ^{174}Yb is accurate enough to be compared with the theoretical results available in the literature [10,11]. The theoretical energy of the upper $6s7s\,^1S_0$ state, calculated recently to eight significant digits using the HFR method [11], corresponds to a theoretical frequency of $f_{02}^T = 1\,029\,808.1$ GHz. Thus, our results in Eq. (5) are already two orders of magnitude more accurate than the most accurate calculation. Therefore, our experimental result for the energy of the $6s7s\,^1S_0$ state can provide a reference value for verifying various theoretical calculation methods such as the multiconfiguration Hartree-Fock method in the framework of the Breit-Pauli corrections and HFR method [11]. The measured energy of the $6s7s\,^1S_0$ state of ^{174}Yb from Eq. (5) is 34 350.6810(24) cm^{-1} and it lies between two published theoretical values with much smaller uncertainty [10,11].

Measurement uncertainties can be further reduced in the future by narrowing linewidth of f_{ceo} of the YLFC by stabilizing the comb components with respect to a highly stable optical frequency standard [22] and stabilizing the probe laser frequency to the center of the resonance fluorescence signal obtained from the Yb atomic beam [12] or optically trapped Yb atoms [16] as discussed above.

VI. CONCLUSIONS

In summary, we have measured the absolute frequency of the $6s6P\,^1P_1$–$6s7s\,^1S_0$ transition of ^{174}Yb at 1077.3 nm in a HCL. The frequency was measured to be 278 281 521.4(7.2) MHz with a relative standard uncertainty of 2.6×10^{-8}. In addition, from the reported value of the transition frequency at 398.9 nm, we determined the absolute frequency of the $6s^2\,^1S_0$–$6s7s\,^1S_0$ two-photon transition at 291.1 nm to be 1 029 807 509.2(7.2) MHz with a relative standard uncertainty of 7.0×10^{-9}. Our measurement uncertainties are accurate enough to be compared to those of the theoretical values available in the literature and therefore can provide reference values to test various theoretical calculation methods for the excited-state energy of Yb isotopes [11,14].

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