Tunable frequency-stabilization of UV laser using a Hallow-Cathode Lamp of atomic thallium

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A frequency-stabilized ultraviolet laser system, locked to the thallium resonant transition of 377.5 nm, was demonstrated using a novel bichromatic spectroscopy technique for tuning the zero-crossing laser-lock point. The atomic thallium system is a promising candidate in atomic parity violation and permanent electric dipole moment (EDM) experiment mainly attributing to its large atom number (Z=81), which enlarge APV and EDM effects by Z² [11]. Its strong 6P₁/₂ → 7S₁/₂ resonant transition, at 377.5 nm, permits optical pumping to the metastable state 6P₃/₂ prepared for the nearly closed transition cycle 6P₃/₂(F = 2) ↔ 6D₅/₂(F = 3) for the laser cooling and trapping experiments [12], which can greatly reduce the systematic shifts and the spectral-broadening effects in potential applications of high-resolution spectroscopy. Due to the high melting point (304°C), atomic Tl HCL is versatile to stabilize the 377.5 nm UV laser for the Tl laser cooling experiment.

The experimental apparatus is shown schematically in Fig. 1. It includes three main parts: HCL spectrometer, atomic beam spectrometer and 377.5 nm laser system. The HCL (Hamamatsu L2783-81NE-TL) is a seethrough type with a ring cathode length of 19 mm and a bore diameter of 3 mm. It is filled with 14 Torr neon as buffer gas. The nominal discharge current is 9 mA, corresponding to 490 V voltage drop across the discharge with a 30 kΩ ballast resistor. The 377.5 nm laser system includes a single frequency cw Ti:sapphire laser operated at 755 nm and a frequency doubler based on a traveling-wave ring cavity with a Brewster-angle cutted LBO crystal. Total power of the generated UV light is typically 1 mW. One part (∼300 µW) of the UV light was sent to the Tl atomic beam system for the sub-Doppler fluorescence spectrum as a shift-free frequency reference at 6P₁/₂ → 7S₁/₂. Another part (∼500 µW) was to the bichromatic spectrometer for the frequency stabilization.

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Laser stabilization on atomic or molecular reference transitions is essential to high precision spectroscopy and laser cooling experiment. For the experiments with low vapor pressure samples, a gas cell as a reference requires a high temperature oven or a beam machine. For example, atoms with high melting points [1], such as atomic strontium [2], ytterbium [3] and thallium (Tl) [4], would require a 300-900°C cell temperature to reach a sufficient number density. Hollow-cathode lamp (HCL), which provide high number densities of atoms or molecules in the ground or excited states using electrical discharge, is a versatile and compact setup for such low vapor pressure spices. Various spectroscopy techniques have been implemented using HCLs to stabilize laser frequency with atomic ytterbium [5], calcium [6], and thorium [7]. However, owing to the high buffer gas pressure in the HCL, a shift and broadening of the spectra [8], which are important as a frequency reference, need to be carefully studied and compensated using a frequency correction in many applications.

In our experiment, a 377.5 nm frequency-doubled Ti:Sapphire laser was locked to the Doppler-free saturation spectroscopy of atomic Tl in a HCL using the bichromatic spectroscopy technique [9,10]. To study the pressure shift, an atomic Tl beam system was simultaneously used to observe the fluorescence spectrum as the shift-free reference for comparison with HCL. A simple method of correcting the frequency shift to the transition center was also demonstrated.

Atomic Tl is an attractive element for atomic parity violation (APV) and permanent electric dipole moment (EDM) experiment mainly attributing to its large atom number (Z=81), which enlarge APV and EDM effects by Z² [11]. Its strong 6P₁/₂ → 7S₁/₂ resonant transition, at 377.5 nm, permits optical pumping to the metastable state 6P₃/₂ prepared for the nearly closed transition cycle 6P₃/₂(F = 2) ↔ 6D₅/₂(F = 3) for the laser cooling and trapping experiments [12], which can greatly reduce the systematic shifts and the spectral-broadening effects in potential applications of high-resolution spectroscopy. Due to the high melting point (304°C), atomic Tl HCL is versatile to stabilize the 377.5 nm UV laser for the Tl laser cooling experiment.

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For the atomic beam spectroscopy, the UV light was
modulated at 2 kHz by a mechanical chopper and interacted perpendicularly with an aperture-collimated atomic beam with a 90 mrad divergent angle. The atomic beam apparatus include a stainless chamber with UV AR coated windows and with background pressure of $10^{-5}$ Torr. The atomic beam was generated by heating a bulk of Tl to 420°C. The laser induced fluorescence was detected by a photomultiplier and demodulated using a lock-in amplifier. The details of the experiment can be found in our previous report [4]. A temperature-controlled Fabry-Perot interferometer with a free spectral range of 300 MHz (in 755 nm) was used to diagnose the laser scanning and calibrate the scanned frequency range.

In order to provide a Doppler-free dispersion-like signal for laser stabilization, the dichromatic spectroscopy and the Doppler-free modulation transfer technique are implemented in the HCL spectrometer. In Fig. 1, the UV laser was separated into two pumping beams and a counter-propagating probe beam. The dichromatic spectroscopy technique requires two pump beams with a frequency difference, which was provided using an AOM (Intraaction) driven by a 260 MHz radio-frequency with a 90 kHz amplitude-modulation. The 0th (∼(Intraaction) driven by a 260 MHz radio-frequency with frequency difference, which was provided using an AOM counter-propagating probe beam. The dichromatic UV laser was separated into two pumping beams and implemented in the HCL spectrometer. In Fig. 1, the and the Doppler-free modulation transfer technique are final for laser stabilization, the bichromatic spectroscopy oscillator. PI: proportional-integral DAQ: data acquisition. VCO: voltage-controlled second harmonic generation. AOM: acousto-optic modulators; ABPR, auto-balanced photo receiver. SHG: Figure 2 shows the hyperfine transition $6P_{1/2}, F = 1 \rightarrow 7S_{1/2}, F = 1$ transition of $^{203}$Tl (C) and $^{205}$Tl (D). Gray dots: optogalvanic spectrum in HCL. Blue dots: saturation absorption spectrum in HCL. Black dots: Doppler shift-free laser induced fluorescence spectrum in atomic beam system. There is a frequency shift between the spectra in HCL and atomic system.

Figure 2 shows the hyperfine transition $6P_{1/2}, F = 1 \rightarrow 7S_{1/2}, F = 1$ at 377.5 nm, where the HCL Doppler-free saturation absorption spectrum (blue dots), the Doppler broadened optogalvanic spectrum (gray dots) and the atomic beam fluorescence spectrum (black dots) were recorded simultaneously. The HCL saturation absorption signal was obtained by the modulation transfer technique with only the 1st order pump beam. The atomic beam fluorescence spectrum was fitted by a Voigt function with a residual Doppler width ∼ 70 MHz. In comparison with the atomic fluorescence spectrum, a relative frequency shift of both the HCL saturated absorption peak and optogalvanic peak was found. This 100 MHz red shift (∆fshift shown in the Fig. 2), which occurred on both two isotopes and all the hyperfine transitions, was contributed mainly from the pressure shift induced by Ne buffer gas [13] in HCL. A part of the shift could be contributed to the first order Doppler shift of the atomic beam spectrometer, because of the non-perpendicularity between the atomic beam and the laser beam.

The lock-in amplifier output is the differential signal between the in- and out-phase signals, referenced to the modulation. In this experiment, the resulted signal was the difference of the two saturation dips from the 1st and 0th order pump beams. A simple model of such
A dispersion-like signal can be written as:

\[ S(\omega) = A_0 G(\omega - \omega_0) - A_1 G(\omega - (\omega_0 - \Delta/2)), \]

where \( A_0 \) and \( A_1 \) are the amplitude-modulation coefficients of the pump beams of the 0th and 1st order, respectively. The parameter \( \Delta \) is the rf frequency of the AOM. Because of a high buffer gas pressure in the HCL, the observed single-beam lineshape under the effect of velocity-changing collisions [14] is gaussian \( G(\omega - \omega_0) = \exp(-(\omega - \omega_0)^2/2\sigma^2) \). Hence, the zero-crossing point \( S(\omega_{zero}) = 0 \), where the laser is locked, is at the frequency:

\[ \omega_{zero} = \omega_0 - \frac{\Delta}{4} + \frac{2\sigma^2}{\Delta} \ln\left(\frac{A_0}{A_1}\right) \]

In the case of perfect overlapping of the three beams (two pump beams and one probe beam), \( A_0 = A_1 \) and the zero-crossing point is at \( \omega_{zero} = \omega_0 - \Delta/4 \). While one of the pump beams is slightly mis-aligned or attenuated, a non-equal amplitude modulation \( A_0 \neq A_1 \) results an asymmetrical dispersion-like profile, which has a shifted zero-crossing point. In our experiment, this feature was utilized to tune the frequency of the locked laser by varying the overlapping of the laser beam and compensates the pressure shift in the HCL to set the zero-crossing point on the resonance frequency of the atomic beam. In the Fig. 3, before adjustment (blue dashed-dotted curve) there is an offset \( \sim 30 \) MHz between the zero-crossing point and fluorescence peak. By adjusting the alignment of one of the pump beam (the red solid curve in the inset of Fig. 3), the zero-crossing point was shifted to match the fluorescence peak. The peak-to-peak width of the line was measured to be 1.2 GHz. The slope of the first derivative curve used as the laser-lock signal was 0.52 mV/MHz at the center frequency and the signal to noise ratio was estimated to be 350 at an integration time of 0.1 s.

Figure 4 shows the Allen deviation for the frequency-stabilized Ti:Sa at various integration times. The error signal of the locked laser shown in the inset was used to evaluate the stability of the laser frequency. The frequency stability was 500 kHz at 0.1 s integration time and reached a value of 50 kHz at 10 s. In the inset of Fig. 4, it shows that the laser was locked onto the fluorescence peak to maintain a continuous excitation of the \( 6P_{1/2} \rightarrow 7S_{1/2} \) transition. This stability can be maintained for a time \( > 500 \) sec in typical experimental condition.

In conclusion, we have demonstrated a tunable zero-crossing laser-lock point of dispersion-like signal using bichromatic spectroscopy within a compact Ti HCL. The demonstrated method, in which the zero-crossing point is shifted to compensate the relative frequency shift of the saturation profile and the resonance line, provides a precise locking to an atomic resonance of interest. The frequency stability with a deviation of 0.5 MHz at a 0.1 sec integration time was achieved, which is sufficiently narrow to be used as a compact frequency reference for the future laser cooling experiments of various atomic species, including atomic thallium.

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References

1. K. J. Ross and B. Sonntag, Review of Scientific Instruments 66, 4409 (1995).
2. G. Ferrari, P. Cancio, R. Drullinger, G. Giusfredi, N. Poli, M. Prevedelli, C. Toninelli, and G. M. Tino, Phys. Rev. Lett. **91**, 243002 (2003).
3. C. J. Bowers, D. Budker, S. J. Freedman, G. Gwinner, J. E. Stalnaker, and D. DeMille, Phys. Rev. A **59**, 3513 (1999).
4. T.-L. Chen, I. Fan, H.-C. Chen, C.-Y. Lin, S.-E. Chen, J.-T. Shy, and Y.-W. Liu, Phys. Rev. A **86**, 052524 (2012).
5. J. I. Kim, C. Y. Park, J. Y. Yeom, E. B. Kim, and T. H. Yoon, Opt. Lett. **28**, 245 (2003).
6. U. Dammalapati, I. Norris, and E. Riis, Journal of Physics B: Atomic, Molecular and Optical Physics **42**, 165001 (2009).
7. W. DeGraffenreid, S. C. Campbell, and C. J. Sansonetti, J. Opt. Soc. Am. B **29**, 1580 (2012).
8. T. Masaki, Y. Adachi, and C. Hirose, Appl. Spectrosc. **42**, 54 (1988).
9. E. van Ooijen, G. Katgert, and P. van der Straten, Applied Physics B: Lasers and Optics **79**, 57 (2004).
10. C.-C. Chou, T. Lin, P.-C. Huang, and M.-H. Chien, Photonics Technology Letters, IEEE **16**, 1948 (2004.).
11. J. Ginges and V. Flambaum, Physics Reports **397**, 63 (2004).
12. I. Fan, T.-L. Chen, Y.-S. Liu, Y.-H. Lien, J.-T. Shy, and Y.-W. Liu, Phys. Rev. A **84**, 042504 (2011).
13. R. S. Dygdaa, R. Bobkowski, and E. Lisicki, Journal of Physics B: Atomic, Molecular and Optical Physics **22**, 1563 (1989).
14. W. H. Richardson, L. Maleki, and E. Garmire, Phys. Rev. A **36**, 5713 (1987).