Prospects of laser cooling in atomic thallium

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(Received 21 August 2011; published 7 October 2011)

One of the most precisely determined upper limits for the electron electric dipole moment (EDM) is set by the thallium (Tl) atomic beam experiment. One way to enhance the sensitivity of the atomic beam setup is to laser cool the Tl atoms to reduce the EDM-like phase caused by the $\mathbf{E}\times\mathbf{v}$ effect. In this report, a cooling scheme based on the $6P_{3/2}(F=2) \leftrightarrow 6D_{5/2}(F'=3)$ transition in Tl is proposed. The absolute frequency measurement of this nearly closed-cycle transition was performed in an atomic beam apparatus. Two Ti:sapphire lasers were frequency-doubled using enhancement cavities in X-type configurations to provide the needed 377- and 352-nm light sources for the optical pumping and cooling transitions, respectively. The absolute frequency of this cooling transition is determined to be 851 634 646(56) MHz.

DOI: 10.1103/PhysRevA.84.042504 PACS number(s): 31.30.jn, 07.57.−c, 37.10.De, 42.62.Fi

I. INTRODUCTION

The Cabibbo-Kobayashi-Maskawa (CKM) phase of the standard model (SM) can account for the *CP* violation observed in the neutral kaon and *B*-meson decays. However, it cannot explain the large baryon number generated in the early universe. This suggests the presence of new sources of *CP* violation. One potential source could stem from the flavor-diagonal *CP* violation, which manifests itself as the coupling between a spin to the external electric field, i.e., the electric dipole moment (EDM). Thus, the existence of EDM in nucleons, atoms, or molecules could stand as proof of the broken symmetries in *CP* and lay the foundation for theories beyond the SM.

Experimental searches for the EDM have returned null results in paramagnetic atoms [1,2], diamagnetic atoms [3,4], muons [5], tauons [6], neutrons [7], and polar molecules [8–10]. Recent reviews on EDM can be found in Refs. [11,12]. One of the most precisely measured upper limits for the electron EDM in the paramagnetic atom system is $d_e < 1.6 \times 10^{-27}$ e cm (90% confidence) set by the ^{205}Tl thermal atomic beam experiment [1]. (N.B.: The most precise electron EDM measurement done to date is from the YbF polar molecule yielding $d_e < 10.5 \times 10^{-28}$ e cm (90% confidence) [10].) The high sensitivity of the current experimental resolution is approaching a level to impose a stringent test on the extended theories of SM. For example, the split supersymmetry (SUSY) model predicts d_e to be $\approx 10^{-30}$–10^{-28} e cm [13]. The main source of systematic errors in the state-of-the-art Tl atomic beam experiment came from the motional magnetic field, $\mathbf{E}\times\mathbf{v}$, induced by the component of the atom velocity that was perpendicular to the applied electric field [1,14]. This systematic effect was partially compensated by a counterpropagating pair of atomic beams. However, a residual EDM-like phase of $\delta = -0.9 \pm 0.8$ (10^{-7}) was still the major uncertainty in the system. To gain a tighter control of this systematic effect, and thereby increasing the EDM measurement sensitivity, it would be necessary to manipulate the velocity of the moving Tl atoms, a realm that calls for the well-established laser cooling technique [15,16].

EDM searches in a laser-cooled system or its variants (e.g., magneto-optical trap, atomic fountain, or three-dimensional optical lattice) have been recently proposed for ^{225}Ra [17,18], ^{133}Cs [19], and ^{171}Yb [20]. In particular, a laser-cooled EDM search was carried out on a Cs atomic beam slowed to 3 m/s [2]. However, the resultant electron EDM of $\approx 10^{-22}$ e cm from this proof-of-principle run was still far from the current upper limit and even farther from the sensitivity expected ($d_e = 1.3 \times 10^{-29}$ e cm) by adopting the laser-cooling scheme in Cs.

The reason to adopt a *heavy* paramagnetic atom such as Tl or Cs is that the atomic EDM can be enhanced by a factor R in comparison to the intrinsic electron EDM. For the ground state of Tl, R is approximately -582 (2.7 times bigger than that of Cs) [21,22]. Thallium is part of the group III elements with its energy levels shown in Fig. 1. Group III contains elements of significant technological importance such as indium and gallium used in modern III-V semiconductor devices. Laser cooling has been achieved on some of the group III elements, including aluminum [23], gallium [24], and indium [25]. For Tl atoms, however, a similar cooling mechanism has not been attempted so far because an appropriate cooling cycle is not easily accessible.

Among the lower lying energy states the only transition that appears practical to be used as a cooling cycle is the $6P_{3/2}(F = 2) \leftrightarrow 6D_{5/2}(F' = 3)$ transition, i.e., "A" and "D" lines in Fig. 1. It is a transition with the highest relative intensity [26] and can be considered as a nearly closed two-level cycle. The allowed decay channels for the $6D_{5/2}(F' = 3)$ excited state are the $7P_{3/2}(F = 2)$ and the $6P_{3/2}(F = 2)$ states. The transition to the $7P_{3/2}(F = 2)$ state becomes a leakage of the cooling cycle. Estimating from the established oscillator strength [27], this leakage accounts for 0.074% of the cooling transition. The subsequent decay of the leakage is to the $7S_{1/2}(F = 1)$ state, followed by four decay destinations: (a) $6P_{3/2}(F = 2)$, (b) $6P_{1/2}(F = 1)$, (c) $6P_{3/2}(F = 1)$, and (d) $6P_{1/2}(F = 0)$. Channel (a) brings the atom back to the cooling cycle. Channel (b) can be repumped by the optical pumping beam (see later sections). Channel (c), whose transition frequency is merely 150 MHz away from the cooling transition, can be "plugged up" by using an acousto-optic modulator frequency shifter to the cooling laser. Channel (d) is the only significant channel that accounts for <0.01% leakage. It is worth noting that

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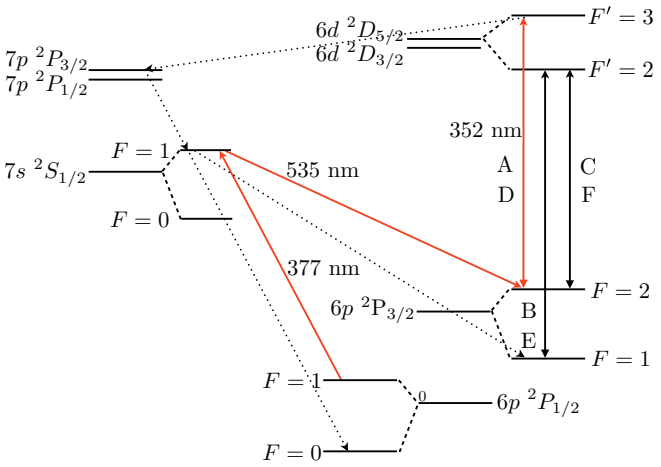

FIG. 1. (Color online) Energy levels of a Tl atom, showing the ground state and the first few excited states. The "A", "B", and "C" hyperfine transition lines are for ^{205}Tl. The "D", "E", and "F" hyperfine transition lines are for ^{203}Tl.

similar repumping schemes (from the metastable state) have been shown to be necessary for the efficient cooling in other atomic systems, such as ^{138}Ba [28] and ^{225}Ra atoms [29].

To access the aforementioned cooling cycle, the Tl atoms have to be first prepared in the $6P_{3/2}(F = 2)$ state. Because its transition to the $6P_{1/2}$ ground state is $E1$ forbidden, $6P_{3/2}$ is a metastable state with a relatively long lifetime (≈0.16 s). Ground-state Tl $[6P_{1/2}(F = 1)]$ can be optically pumped to the metastable state via the $7S_{1/2}(F = 1)$ excited state [30]. The branching ratio from the $7S_{1/2}$ state to the $6P_{1/2}$ and the $6P_{3/2}$ states is about 1:2.[1] The hyperfine splitting of the $6P_{1/2}$ state is 21.1 GHz corresponding to a temperature of 1.01 K. This implies that 50% of the ground-state electrons will be thermally populated to the $F = 1$ state at room temperatures prior to the optical pumping. Our calculation shows that more than 33% of atoms can be pumped to the $6P_{3/2}(F = 2)$ metastate within 5 μs.

In this article, we demonstrate the apparatus to access the proposed cooling route in Tl, namely, the $6P_{3/2}(F = 2) \leftrightarrow 6D_{5/2}(F' = 3)$ transition, and perform an absolute frequency measurement for this transition with an optical frequency comb in an atomic beam setup. A similar frequency characterization scheme was previously applied for the Cs atomic beam [31]. Aside from contributing to the experimental progress toward the EDM search in laser-cooled Tl, the resultant hyperfine structures reported in this article may be used for testing the atomic wave functions used in parity-nonconservation calculations [32]. The layout of the paper is as follows. First, the Tl atomic beam apparatus is described in Sec. II followed by details about our optical layout in Sec. III and our results and discussion in Sec. IV. The conclusion is given in Sec. V.

[1]This is estimated from the lifetimes of the $7S_{1/2}$ state. The lifetime corresponding to the $7S_{1/2} \leftrightarrow 6P_{1/2}$ transition is 23 ns. The lifetime corresponding to the $7S_{1/2} \leftrightarrow 6P_{3/2}$ transition is 12 ns.

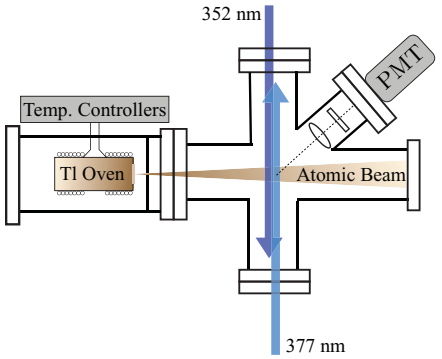

FIG. 2. (Color online) Schematic of the main chamber where the counterpropagating optical pumping (377 nm) and cooling laser beams (352 nm) intersect with the collimated Tl atomic beam. A photomultiplier tube (PMT) is used to collect the fluorescence signal.

II. THERMAL ATOMIC BEAM OF Tl

The atomic beam apparatus used in this experiment is shown in Fig. 2. Thallium granules (99.999%, Strem Chemicals 81-0055) were placed in an oven which was heated by two sets of resistive coils and contained a 2-mm aperture on one side. One coil set was wound at the "back" end of the oven near the vacuum pumps while the other set was wound at the "front" end of the oven to keep the aperture at elevated temperatures to avoid clogging. The current used to heat the coils was limited to 3 A by two independent home-made temperature controllers. The background pressure in the oven was ≈10^{-5} Torr, maintained by a turbo (170 l/s) and rotary pump combination. The typical oven temperature used to generate the atomic beam for this experiment was 450 °C (v = 243 m/s). A 2-mm aperture was placed 4 cm away from the oven to narrow the distribution of the atomic velocities. The collimation ratio was geometrically determined to be 0.05, corresponding to an atomic beam divergence θ of 2.86°.[2] It was estimated that this velocity narrowing by apertures should yield a theoretical reduction in the Doppler broadening from 1 GHz (uncollimated beam) to 58 MHz (aperture collimated beam) with a residual transverse velocity of 12.2 m/s.[3] This reduced Doppler width is comparable to the natural linewidth Γ of the $6P_{3/2} \leftrightarrow 6D_{5/2}$ transition of 23 MHz.[4] Behind the second aperture was the interaction chamber where the atomic beam and two counterpropagating laser beams (377 and 352 nm) intersected. This interaction chamber was heat insulated by a water-cooled flange from the oven and was connected to another turbo pump (50 l/s) for maintaining

[2]Let the size of the second aperture be b and the distance from it to the oven aperture d, then the collimation ratio is $\frac{b/2}{d}$. This estimate is only valid if the oven aperture can be approximated as a point source, i.e., the mean free path ($\lambda = \frac{RT}{\sqrt{2}\pi r^2 N_A P}$, where r is the diameter of a Tl atom) is much longer than d. The mean free path λ is ≈ 170 m in this experiment.

[3]The velocity can be calculated by $v_p = \frac{\omega_D c}{\omega_0 2\sqrt{\ln 2}}$, where ω_0 is the absolute frequency of the "A" line.

[4]The natural linewidth can be estimated from the uncertainty principle ($\Delta E \Delta t = h/2\pi$).The lifetime of the upper state is ≈7 ns.

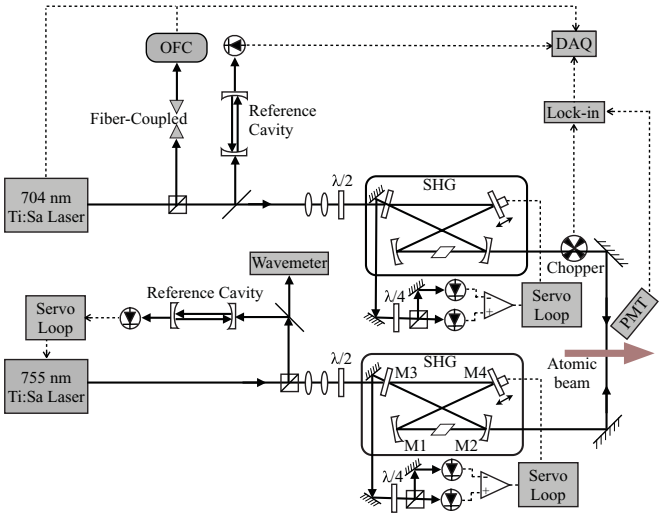

FIG. 3. (Color online) Optical layout of this experiment. Refer to the main text for detailed functions of the components.

pressures below 10^{-5} Torr. All view ports on the interaction chamber were made of fused silica with antireflective coatings in the UV range.

III. LASER SYSTEMS

The optical layout of this experiment is shown in Fig. 3. Two light sources were needed. The 377-nm (≈0.2-mW) optical pumping source for the $6P_{1/2} \rightarrow 7S_{1/2} \rightarrow 6P_{3/2}$ transition was obtained by frequency-doubling a Ti:sapphire laser (Coherent, MBR-110) locked to a temperature-stabilized reference cavity (the free spectral range is 305 MHz) using a servo loop. The 352-nm (≈10-μW) source for the $6P_{3/2} \leftrightarrow 6D_{5/2}$ laser-cooling transition was obtained by frequency-doubling another Ti:sapphire laser (TekhnoScan, TIS-SF-07). The intensity of this laser was partially branched off to an optical frequency comb (OFC) for the absolute frequency measurement and to another reference cavity for the frequency calibration of hyperfine measurements. The low power output of the 352-nm light was due to the low emission intensity of the Ti:sapphire pumping source near 704 nm.

The second harmonic generation cavities were both in an X-type traveling-wave ring configuration [33]. The nonlinear crystals were Brewster cut Lithium Triborate (Castech, $3 \times 3 \times 10$ mm^3) mounted on a multiaxis adjustable stage. The astigmatism caused by the Brewster-angled crystal was compensated by a "negative astigmatism" generated by the off-axis reflections between two curved (the radius of curvature is 10 cm) mirrors (see Fig. 3). The mirror "M2" also functioned as the output coupler. The round-trip power loss of our cavity was 3%. Therefore, the impedance matching condition required for the maximum enhancement effect was achieved by using a 3% flat input coupler "M3" ($T = 3\% \pm 0.5\%$). A thin plane mirror "M4" was mounted on a piezoelectric transducer permitting a cavity length adjustment on the order of GHz for the frequency locking. The cavity is locked to a dispersion-shaped resonance using the Hänsch-Couillaud locking scheme [34].

The florescence signal (30 s/scan) was collected by a photomultiplier tube (Electron Tubes 9111B) through a $f = 30$ mm lens and a 352-nm bandpass filter (Thorlabs FB350-10) and was demodulated using a lock-in amplifier. The modulation frequency of the chopper was 2 kHz on the 352-nm laser beam. The laser beams were collimated to a beam size of 3×10 mm^2 with the long axis parallel to the atomic beam. The optical pump 377-nm laser propagated in the opposite direction with the same beam size, but without modulation. The intensity noises of both SHG light sources caused by kHz acoustic vibrations of the ambient were <5%.

The frequency comb used in this experiment was similar to our previous work [35]. In short, a mode-locked Ti:sapphire ring cavity (Gigaoptics, Gigajet 20) was pumped by a 5-W frequency-doubling Nd:YVO_4 laser at 532 nm (Spectra-Physics, Millennia V) to generate <50-fs pulses with a $f_r = 1$ GHz repetition rate. Supercontinuum comb lines spanning an octave were obtained from the femtosecond pulses using a photonic crystal fiber (OFS Lab). A stable microwave source, consisting of a Rb clock (SRS, PRS10), a low-noise quartz oscillator, and a global positioning system receiver, was used as the frequency reference for all of the radio-frequency electronics, including the synthesizers, analyzers, and counters. The offset frequency f_o was detected by the interferometric self-referencing method [36,37]. With a gate time of 1 s, the root-mean-square fluctuations in f_r and f_o were 3.63 and 11 mHz, respectively. The accuracy of our frequency comb system was verified by measuring the a_{10} line of the $^{127}I_2$ molecular iodine R(56)32-0 transition (≈532 nm) [38]. Our measurement result was in agreement with the international standard for this transition with a <5-kHz deviation.

IV. RESULTS AND DISCUSSION

The optical pumping source was set to the peak center of the $6P_{1/2}(F = 1) \rightarrow 7S_{1/2}(F' = 1)$ transition (see Fig. 4). It also served as an isotope selector by setting the frequency on different peak centers. The frequency jitter of the locked optical pumping source was ≈15 MHz, limited by the linewidth of the resonance peaks of the reference cavity.

Typical spectra of the $6P_{3/2} \leftrightarrow 6D_{5/2}$ transitions of ^{205}Tl and ^{203}Tl are shown in Fig. 5. The lower signal-to-noise

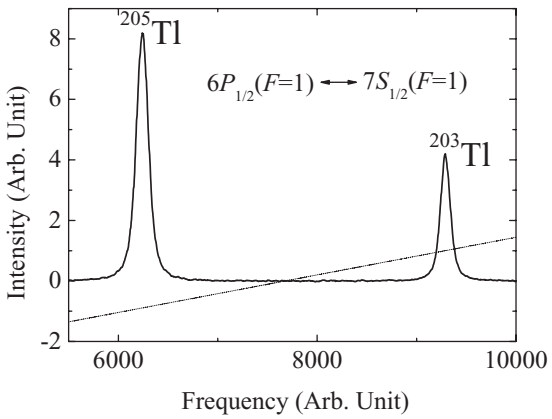

FIG. 4. Optical pumping transition for the two isotopes. The straight line represents the frequency ramping signal of the laser. The PMT was biased at 0.7 kV and the lock-in amplifier settings were as follows: 10-mV sensitivity, $T_c = 10$ ms/12 dB, $\theta = 159.98°$, and $f_{\text{ref}} = 2$ kHz. The oven pressure (450 °C) was 4.1×10^{-7} Torr.

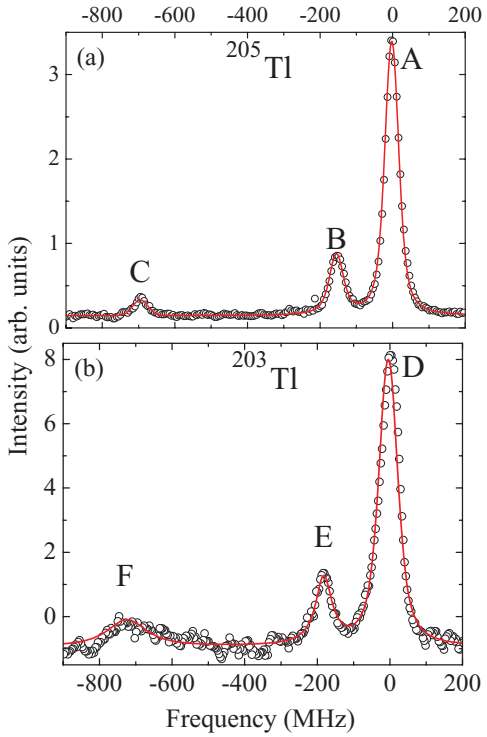

FIG. 5. (Color online) The $6P_{3/2} \leftrightarrow 6D_{5/2}$ hyperfine spectra of (a) ^{205}Tl, with A ($F = 2 \rightarrow F' = 3$), B ($F = 1 \rightarrow F' = 2$), and C ($F = 2 \rightarrow F' = 2$) lines, and (b) ^{203}Tl with D ($F = 2 \rightarrow F' = 3$), E ($F = 1 \rightarrow F' = 2$), and F ($F = 2 \rightarrow F' = 2$) lines. Solid lines are the fits; circles are the data points.

ratio of the ^{203}Tl measurement was attributed to the lower isotopic abundance (the naturally occurring Tl has 30% ^{203}Tl and 70% ^{205}Tl). The "zero" frequency was set on the centers of the $F = 2 \leftrightarrow F' = 3$ transition for both isotopes. The fitted full width at half maximum was 25(4) MHz for the ^{205}Tl "A" line and 33(1) MHz for the ^{203}Tl "D" line. Both values were comparable to the natural linewidth (23 MHz) of the corresponding transitions. The collimating slits were unlikely to reduce the Doppler broadening to such a degree according to our earlier estimation (Sec. II). The virtually non-Doppler-broadened linewidth was due to the preselection of the atomic velocity group by the optical pumping process, also known as the "line-narrowing" effect [44]. A "frequency shift" could also occur in the measured absorption line if there is a nonperpendicular intersection between the laser beams and the atomic beam. However, assuming that the degree of this misalignment is no more than the divergence of the atomic beam, we estimate that this shift was approximately 29 MHz.

Our absolute frequency measurement for the ^{205}Tl "A" line (the laser-cooling transition) yielded

$$f(6P_{3/2}(F = 2) \leftrightarrow 6D_{5/2}(F' = 3)) = 851\,634\,646(56)\ \text{MHz},$$

in which the main source of uncertainty was coming from the jittering of our laser system and the counting error due to the weak beating signal between the Ti:sapphire laser and the frequency comb. Our fundamental laser (702 nm) had a jitter about 7 MHz in 1 ms and exhibited a low power distribution near the 705-nm spectral range. It is also worth pointing out that the other fine-structure transition ($6P_{3/2} \leftrightarrow 6D_{3/2}$) has not been observed within the expected window (849 140–849 190 GHz). This could be attributed to its weaker transition probability which was 17% of the transition probability of the $6P_{3/2} \leftrightarrow 6D_{5/2}$ transition [27].

To deduce the separations of the hyperfine transitions, the spectra were fitted with three Voigt functions using ROOT.[5] The resulting hyperfine splitting for $6D_{5/2}$ in ^{205}Tl was 693(18) MHz, which was within uncertainties of the previous measurement, 680.7(9) MHz, derived from the corresponding hyperfine constant [40]. On the other hand, our hyperfine splitting for $6D_{5/2}$ in ^{203}Tl was 718(74) MHz. This was significantly different from the literature value of 680.7(9) MHz [40]; although, they still agreed with each other within our large error bar. The precision of our hyperfine measurement in ^{203}Tl was limited by the weak "F" line, which was poorly resolved [Fig. 5(b)] due to its intrinsically low transition probability and the low natural abundance of ^{203}Tl in our atomic beam.

The hyperfine structures for the $6P_{3/2}$ state were also analyzed in order to obtain an estimate of its isotopic hyperfine anomaly. The hyperfine splittings of $6P_{3/2}$ are 534(74) and 540(20) MHz for ^{203}Tl and ^{205}Tl, respectively. The isotopic hyperfine anomaly was deduced to be $\Delta = \frac{A_1 g_2}{A_2 g_1} - 1 = -14 \times 10^{-4}$, where g_1 and g_2 are the gyromagnetic ratios of the two isotopes [45]. This value, obtained via our all-optical experiment, could be compared with the the same hyperfine anomaly measured by the magnetic resonance method, which is $-16.26(2) \times 10^{-4}$ [39]. The accuracy in the $6P_{3/2}$ hyperfine anomaly was also constrained by the weak "F" line in ^{203}Tl. The hyperfine constants of $6D_{5/2}$ and $6P_{3/2}$ reported in this experiment are summarized in Table I along with existing literature values.

[5]A data analysis tool developed at CERN. It can be downloaded from "http://root.cern.ch/drupal/".

TABLE I. Summary of hyperfine constants measured in this work and previously reported values (in MHz).

	$6P_{3/2}$ (^{203}Tl)	$6P_{3/2}$ (^{205}Tl)	$6D_{5/2}$ (^{203}Tl)	$6D_{5/2}$ (^{205}Tl)
This work	267(37)	270(10)	239(25)	231(6)
Ref. [39]	262.031(1)	265.0383(1)		
Ref. [40]			226.9(3)	228.9(3)
Ref. [41]	262.1(0)	265.1(0)		
Theory [42]		248		183
Theory [43]		353		215.4

The cooling force (or radiation-pressure force) exerted on the Tl atoms can be calculated by multiplying the photon absorbing rate with the momentum k carried by each photon. However, due to the finite (<0.01%, see Sec. I) leakage channel via $7P_{3/2}$, the cooling photon absorbing rate cannot go to infinity even under a continuous exposure to the cooling beam. We estimate that *on average* there will be 10^4 absorption events before the leakage to the $6P_{1/2}(F=0)$ ground state occurs. With an additional repumping for the $6P_{3/2}(F=1) \leftrightarrow 6D_{5/2}(F'=2)$ transition, 10^4 absorption events correspond to a 70-μs interaction time, since each cooling cycle takes 7 ns [46]. The typical laser-cooling intensity needed is a fraction of the saturation intensity. For the $6P_{3/2} \leftrightarrow 6D_{5/2}$ cooling transition, the saturation intensity is 22 mW/mm^2 ($I_0 = \frac{4\pi^2 hc}{\lambda^3} A$, where A is the Einstein coefficient). The cooling power available in our system (10 μW, limited by the weak emission of the Ti:sapphire laser around 704 nm) is for the moment too low to perform such a laser-cooling experiment on the Tl atomic beam. If a cooling power $\frac{I}{I_0} = 0.5$ is available, then the transverse and longitudinal components of the atomic beam can be slowed to 4 m/s (200 mK) and 80.1 m/s (79 K), respectively, assuming the detuning is Γ.

V. CONCLUSION

A promising laser-cooling scheme in Tl, namely, $6P_{3/2}(F=2) \leftrightarrow 6D_{5/2}(F'=3)$, was proposed and evaluated. The atomic beam apparatus and light sources to access the laser-cooling transition were demonstrated. The absolute frequency of the cooling transition was measured by a self-referencing frequency comb. Hyperfine structures of $6D_{5/2}$ and $6P_{3/2}$ were also analyzed and they may be useful for the precision determination of the atomic wave functions in the electroweak theory and for the parity-nonconservation measurement in atomic Tl. A laser cooling of Tl atoms has not been achieved by our experiment due to the low cooling power. Recent development of a UV light source based on the external cavity diode laser is a favorable candidate that can lead toward the ultimate realization of laser cooling of Tl atoms.

ACKNOWLEDGMENTS

This work was supported by the National Science Council of Taiwan under Grants No. 97-2112-M-007-004-MY3 and 96-2112-M-007-014-MY3.

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