Frequency stabilization of the frequency doubled DOFA to the ^{127}I$_2$ line for calcium spectroscopy

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Frequency doubled diode oscillator and fiber amplifier (DOFA) is frequency-stabilized to a hyperfine line of molecular iodine for calcium spectroscopy. The frequency doubling of DOFA is demonstrated using an Mg-doped periodically poled stoichiometric lithium tantalate crystal for generating a 544-nm beam. Saturated absorption spectroscopy is performed on the molecular iodine at 544 nm to find a hyperfine line for stabilizing the 272-nm beam to the calcium-48 transition. Stabilization of the 272-nm beam using the stabilization of the 544-nm beam to the corresponding ^{127}I$_2$ line is discussed.

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Frequency stabilization has been an important technique for many applications, such as a trace analysis, neutral atom trap, optical pumping, and electromagnetically induced transparency[1−3]. In general, frequency stabilization has been performed on a hyperfine level of target atomic and molecular species for the precise control of the laser frequency. However, the vapor densities of many metallic elements cannot be maintained stably for a long experimental period due to a short lifetime coming from condensation on the surface. Moreover, in the case of spectroscopy on low abundant metallic atom, such as calcium-48 and trace-elements, it is not easy to stabilize the frequency to the corresponding transition due to a poor signal-to-noise ratio (SNR). Recently, frequency stabilization using ^{127}I$_2$ has been reported instead of direct frequency locking to the atomic transition in the visible region to overcome the above difficulties[4]. Molecular iodine has many absorption lines over 500 nm, and thus it is used for frequency references in visible and infrared regions[5].

We consider the $4s^2$ $^1S_0$–$4s5p$ $^1P_1$ transition of calcium-48 for the frequency stabilization. A Ca-48 isotope can be used as a raw material for the measurement of neutrinoless double beta decay and an ion beam source for searching new heavy elements in an accelerator[6,7]. Frequency stabilization to the $4s^2$ $^1S_0$–$4s5p$ $^1P_1$ transition can be used for an isotope analysis and calcium isotope production. However, the natural abundance of calcium-48 is around 0.2%, and the resonant wavelength corresponding transition is around 272 nm. In addition, it is difficult to produce stable calcium vapor due to the calcium-coating on the cell windows and a fast loss of atoms[8]. In this work, we introduce a frequency stabilization technique of an ultraviolet beam to a rare abundant metallic atom transition in the ultraviolet region.

For this work, we should consider the 272-nm beam generation as a first step. The 272-nm beam is not easy to be generated using a dye laser or a Ti:sapphire laser owing to the complexity and hard maintenance of the system. Recently, diode oscillator and fiber amplifier (DOFA) has been developed for a wavelength longer than $1\,\mu m$ and has several advantages, such as compactness, reliability, good beam quality, and tunability even under high power operation. Thus, we employed a DOFA operated at 1 088 nm and double stages of frequency doubling for continuous-wave single-frequency tunable 544 nm and 272 nm beams. The frequency conversion stages consisted of a single-pass second harmonic generation (SHG) stage with a Mg-doped periodically poled stoichiometric LiTaO$_3$ (PPsLT) crystal for a 544-nm beam, and a commercial ring-cavity frequency doubler for a 272-nm beam. Therefore, we investigated the frequency doubling with a quasi-phase matched crystal for our application.

To stabilize the 272 nm beam to the $4s^2$ $^1S_0$–$4s5p$ $^1P_1$ transition of calcium-48, we proposed a stabilization of the frequency of the 2nd harmonic beam to the molecular iodine lines instead of the direct stabilization of the 4th harmonic beam to the calcium-48 transition. We need to find the nearest molecular iodine line at around 544 nm while the frequency quadrupled DOFA matches the calcium-48 transition.

In this letter, we present a frequency doubled DOFA with a quasi-phase matched crystal for a continuous-wave (CW) single-frequency tunable 544-nm beam. We also present a demonstration of the saturated absorption spectroscopy on molecular iodine at around 544 nm. Next, we compare a hyperfine line of molecular iodine at 544 nm with a calcium transition at 272 nm. We also discuss the compensation of the frequency difference and frequency stabilization of a 544-nm beam while the 4th harmonic beam is resonant to the calcium-48 transition.

The system setup is shown in Fig. 1(a). The light source consists of a DOFA, a 2nd harmonic generation stage with the Mg-doped PPsLT crystal in a single-pass configuration, and a commercial 4th harmonic generator (wavetrain, Spectra physics) with BBO crystal. The ex-

ternal cavity diode laser (ECDL) with the power of 5 mW at 1 088 nm was seeded to an ytterbium doped fiber amplifier (Keopsys Inc.) with a polarization maintaining fiber. The power was amplified up to around 10 W maintaining single frequency and polarization. The amplified beam was collimated and went through an optical isolator to prevent damage by back reflection.

The 1 088-nm beam was focused on the Mg-doped PPsLT crystal (length: 30 mm, thickness: 0.5 mm) in a heat oven for the 2nd harmonic generation. The temperature of the heat oven was maintained at around 67 °C for the optimum phase matching. The 544-nm beam was separated from the mixed beams of the 544 and 1 088-nm beams by two dichroic mirrors and the rest of the 1 088-nm beam was blocked. The 544-nm beam was split into two parts: one was used for the saturated absorption spectroscopy on molecular iodine and the other was used for the 4th harmonic generation for the calcium spectroscopy. The setup of the saturated absorption spectroscopy is depicted in Fig. 1(b). The laser beam power for the saturated absorption spectroscopy was controlled by the set of a half-waveplate and a polarizing beam splitter, but is not shown in the figure. The 544-nm beam for the spectroscopy went through an acousto-optic modulator for the frequency shift to compensate the frequency difference for matching the hyperfine state signal of the molecular iodine in the visible region, and the resonance signal of calcium atoms in the ultraviolet region. The frequency shifted beam was split into three beams, a reference beam, probe beam, and pump beam as shown in Fig. 1(b). The reference beam was used for compensating the Doppler broadened absorption signal and improving the SNR. The dispersive curve was obtained by modulating the shifting frequency of the acousto-optic modulator, and was used as a feedback signal for the frequency stabilization. The length of the iodine cell was around 100 mm, and the iodine cell was operated at room temperature.
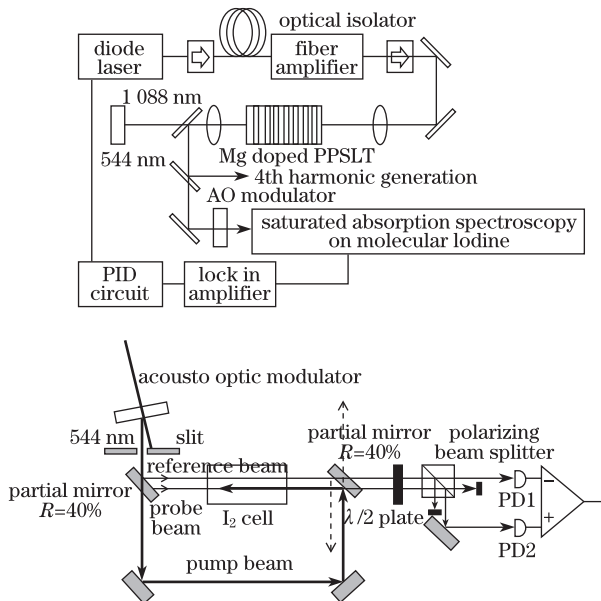

Fig. 1. (a) Experimental setup for the diode oscillator and fiber amplifier and its frequency stabilization; (b) saturated absorption spectroscopy on molecular iodine using the 544-nm beam.
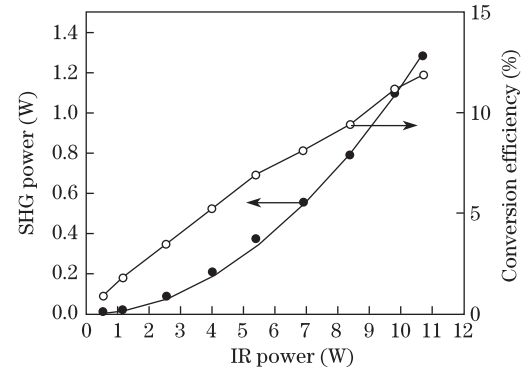

Fig. 2. The second harmonic generation power versus the DOFA power. The wavelengths of the fundamental beam and SHG beam are around 1 088 and 544 nm, respectively.

The 544-nm beam was delivered to the 4th harmonic generator for producing a 272-nm beam, and the 272-nm beam was used for the calcium spectroscopy.

As mentioned above, the 544-nm beam was produced by the second harmonic generation of the DOFA with the Mg-doped PPsLT crystal in a single pass configuration. The conversion efficiency was optimized by varying the confocal parameters, oven temperature, and position of the focus along the beam direction. The optimized beam waist in the focus was around 40 μm. Figure 2 shows the frequency doubling result versus the fundamental power under optimum condition. The power of the 544-nm beam (solid circle) reached up to 1.3 W at around 10.5 W of the 1088 nm beam. The conversion efficiency was around 12% at maximum SHG power. The data were fitted well with a square function (solid line). The power depletion and thermal effect were not observed at this power level. The beam profile of the 544-nm beam was Gaussian, but the shape was asymmetric due to the astigmatism. The single longitudinal mode operation was checked by the confocal Fabry Perot etalon while the diode laser was scanned.

The frequency quadrupling was performed using a commercial external ring frequency doubler (wavetrain, Spectra physics). The 544-nm beam went through the mode matching optics and was incident on the enhancement cavity for the generation of the 272-nm beam. The astigmatism was compensated by tilting the mode matching lenses. A BBO crystal with a length of 10 mm was used in the cavity. The 272-nm beam reached up to around 20 mW and it was provided for the resonant ionization of calcium atoms in a time-of-flight mass spectrometer.

We considered the P88(27-0) and R59(29-1) lines of the molecular iodine using an atomic database and also performed saturated absorption spectroscopy on these two lines by using 544-nm beam. Figure 3 shows the saturated absorption signal of the molecular iodine while the 544-nm beam was scanned. The Fabry Perot etalon signal shows the single frequency operation in the visible region. The result shows that two P88(27-0) and R59(29-1) lines were overlapped, and they were identified using the database. The experiment was performed at pump and probe beam powers of about 8 and 1 mW, respectively. The beam diameters were around 2 mm. At this condition, we observed 26 resolved lines with a linewidth
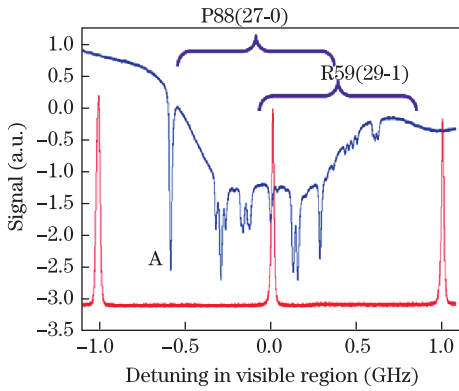
Fig. 3. Hyperfine structure of the molecular iodine at 544 nm and the Fabry Perot etalon signal. The signal at A was considered in frequency stabilization.
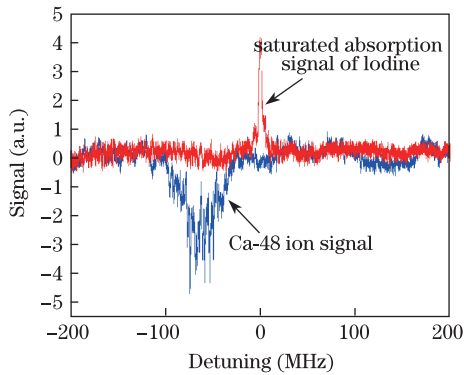

Fig. 4. Hyperfine line of molecular iodine at 544 nm and the resonance line of Ca-48 at 272 nm. The x axis represents the frequency detuning of the 544 nm beam from the molecular iodine signal.

of around 16 MHz. Varying the pump power from 2 to 8 mW, the signal width broadened from 6 MHz to 16 MHz owing to the power broadening.

In this experiment, the Doppler background of the signal was compensated by the dual photodiodes. The background was completely compensated by the subtraction of the reference signal at the far-resonant condition, but was not at the resonant condition because the linear absorption profile was affected by the strong pump radiation.

Next, we found the nearest hyperfine line to the corresponding calcium-48 transition[5,9]. For this, we performed saturated absorption spectroscopy on the iodine molecules using the 2nd harmonic beam (544 nm) and the resonant ionization spectroscopy on calcium using the 4th harmonic beam (272 nm), simultaneously (Fig. 4). The 272-nm beam was provided to optical-pump the ground state calcium to the metastable state, and a strong pulse laser was employed to ionize the metastable state calcium. The photo-ions were generated when the 272-nm beam was resonant on the calcium-48 transition. The ions were measured by a multichannel plate as shown in Fig. 4. Figure 4 shows the saturated absorption signal with the 2nd harmonic beam (544 nm) and the ionization signal of calcium-48 with the 4th harmonic beam (272 nm), simultaneously. The saturated

absorption signal in the figure was the first hyperfine signal with mark A in Fig. 3. The axis represents the detuning of the 544-nm beam from the iodine signal. The frequency difference between the iodine signal and the frequency of the SHG beam when the UV beam was located at the peak of the Ca-48 ion signal was measured to be around 65 MHz. The peak position of the Ca-48 ion signal was determined after smoothing the data points. The width of the ion signal was dominantly dependent of the divergence of the calcium atomic beam. The results in Fig. 4 show that the 65-MHz up-shift by the acousto-optic modulator is required to match the hyperfine signal of $^{127}I_2$ at 544 nm with the $4s^2\ ^1S_0$–$4s5p\ ^1P_1$ transition of calcium-48 at 272 nm.

Therefore, the acousto-optic modulator with a carrier frequency operating at 65–95 MHz was employed and operated at 65 MHz. The cavity enhancement technique for the 4th harmonic generation allows only a slow frequency scan for the cavity locking, and thus the wavelength modulation with the acousto-optic modulator was provided to obtain the dispersive curve as shown in Fig. 5(a). The offset of the signal was observed owing to the intensity modulation by the AO modulator. This offset was compensated by the electronic-offset. As a result, the dispersive curve was used as a feedback signal for the ECDL. The shifted 544-nm beam was stabilized to the hyperfine line of the molecular iodine as shown in Fig. 5(b). The leading part shows the dispersive curve signal during manual scanning. The peak-to-peak voltage of the dispersive curve signal is correspondent to 16 MHz, and thus, the frequency drift was measured to be around 1 MHz. At the moment, the 4th harmonic beam of the wavelength was resonant on the calcium-48 transition.
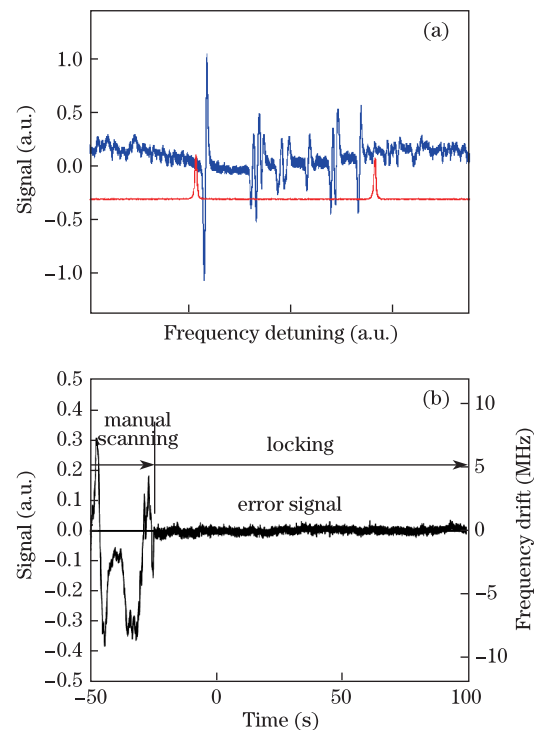

Fig. 5. (a) Dispersive curve by modulating the acousto-optic modulator; (b) error signal for the frequency locking to the first transition (marked A in Fig. 3) of molecular iodine.

In conclusion, we introduce a stable frequency stabilization technique resonant with the calcium-48 transition in the ultraviolet region. At first, we demonstrate a coupled method of a single-pass SHG for 544-nm generation and a cavity enhancement technique for the 272-nm beam. When the frequency-shifted 544-nm beam by 65 MHz is resonant with the iodine hyperfine line, the frequency quadrupled DOFA (272 nm) is resonant with the calcium-48 transition. Therefore, the 272-nm beam is stabilized to the calcium-48 transition by stabilizing the frequency shifted 544-nm beam to the hyperfine line of the molecular iodine. This technique is more advantageous than the direct frequency locking technique in the metallic vapor and is applicable to spectroscopy on rare elements in ultraviolet region.

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