Frequency stabilization of a 399-nm laser by modulation transfer spectroscopy in an ytterbium hollow cathode lamp*

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The modulation transfer spectroscopy in an ytterbium hollow cathode lamp at 399 nm is measured. The error signal for frequency locking is optimized by measuring the dependences of its slope, linewidth and magnitude on various parameters. Under the optimum condition, the laser frequency at 399 nm can be stabilized. The long-term stability of laser frequency is measured by monitoring the fluorescence signal of the ytterbium atomic beam induced by the locked laser. The laser frequency is shown to be tightly locked, and the stabilized laser is successfully applied to the cooling of ytterbium atoms.

Keywords: laser frequency stabilization, modulation transfer spectroscopy, ytterbium, hollow cathode lamp

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1. Introduction

Recent experimental results show that the fractional frequency uncertainty of the strontium optical atomic clock is close to 1×10^{-16},[1] surpassing that of the cesium fountain atomic clock.[2] This means that the optical frequency standard may become the next generation frequency standard. The alkaline-earth-metal atoms as well as ytterbium (Yb) atoms are promising candidates for future optical frequency standards due to the existence of mHz-line-width transitions between their singlet and triplet states. With the help of laser cooling and trapping, especially the optical lattice technique, the optical atomic clocks using various atomic species are being developed at several laboratories around the world.[1,3–6] The stability of the atomic clock depends on the signal-to-noise ratio, and in turn on the number of cold atoms which is limited by the first-stage cooling process. Therefore, producing as many cold atoms as possible in the first-stage cooling is one of the key steps in the optical atomic clock. To achieve this goal, the frequency-stabilized laser must be utilized. A few frequency-locking schemes for this type of atoms have been shown to work well.[7–9] For example, the frequency modulation spectroscopy and saturated absorption spectroscopy have been applied, respectively, to strontium[1,10,11] and ytterbium atoms.[7,8,12]

The 399 nm $^1S_0 - {}^1P_1$ transition of Yb atoms is chosen to realize the first-stage cooling and trapping of Yb atoms. Since the natural linewidth of the $^1S_0 - {}^1P_1$ transition is about 28 MHz, the laser employed for cooling is required to have a linewidth of 1 MHz or less with frequency drift of less than 1 MHz/h. Therefore, the frequency of the cooling laser needs to be locked to the $^1S_0 - {}^1P_1$ transition line of Yb atoms. In the present work, an efficient and simple frequency-locking scheme using the modulation transfer spectroscopy (MTS) technique with a hollow cathode lamp (HCL) is introduced. The optimum parameters for maximizing the MTS signal are also analysed. The MTS employs three advanced techniques: high frequency modulation, frequency modulation (FM) sideband spectroscopy and four-wave mixing (FWM).[13–16] MTS is insensitive to the background absorption and directly provides a dispersion-like signal with a zero-crossing at line centre which is used to lock a laser to a well-defined reference frequency with high sensitivity. Fur-

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thermore, using HCL instead of evacuated heat pipe or vacuum chamber makes the apparatus of MTS simpler.

2. Experimental setup

In our experiment, we choose the ytterbium atom source to realize an Yb optical atomic clock. The $^1S_0$ to $^1P_1$ broad transition ($\Gamma/2\pi = 28$ MHz) at 399 nm is used for the first-stage Doppler cooling. The modulation transfer spectroscopy technique is applied to lock the frequency of the cooling laser at 399 nm. MTS originates from saturation spectroscopy technique and is utilized to detect atomic or molecular transitions with high accuracy. In MTS, a frequency or phase modulated optical pump beam passes through a resonant gaseous medium. An un-modulated probe beam is counter-propagated with the pump beam. With the nonlinear interaction of the two counter-propagating beams within the medium with a sufficiently nonlinear susceptibility, modulation transfer occurs where an un-modulated probe acquires sidebands. This transfer phenomenon is a non-linear four-wave mixing process in the atomic sample which has been described in Refs.[14]–[16].

Figure 1 is the schematic diagram of our modulation transfer spectroscopy setup for the 399 nm laser frequency stabilization. The laser system in dash-dot-line frame is SYST TA-SHG 110 from Toptica Photonics, which is comprised of an external-cavity diode laser (ECDL), a tapered amplifier (TA) and a second-harmonic generator (SHG). The output power of this laser system is about 120 mW at 399 nm, and the laser linewidth is about 1 MHz. In this experiment the laser beam output from SHG is split into two beams by the first polarizing beam splitter (PBS1), one is sent to the Yb atomic beam apparatus, and the other is sent to the frequency stabilization system. The latter is double passed through an acousto-optic modulator (AOM1) having the centre frequency of 80 MHz and the bandwidth of 40 MHz for frequency tuning, then the output beam is split into two beams by the second PBS2, one is for pumping, and another is for probing. The intensity ratio of the probe and pump beams is controlled by the second half-wave plate (HP2), and the polarizations of both beams are independently controlled by half-wave plates (HP3 and HP4) respectively.

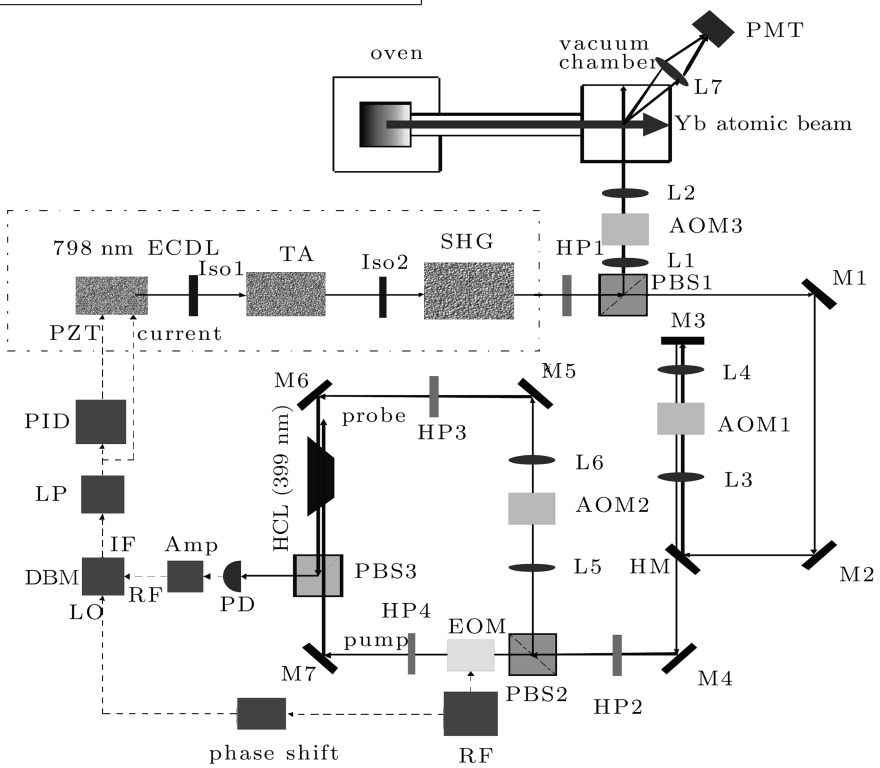


Fig. 1. Schematic diagram of the experimental layout. ECDL, external cavity diode laser; Iso, isolator; TA, tapered amplifier; SHG, second harmonic generation; HP, half-wave plate; PBS, polarizing beam splitter; L, lens; M, mirror; HM, half-mirror; EOM, electro-optic modulator; AOM, acousto-optic modulator; HCL, hollow cathode lamp; DBM, double-balanced mixer; PD, photodiode; PMT, photomultiplier tube; Amp, amplifier; PZT, piezoelectric transducer; LP, low-pass amplifier. The solid lines indicate the light beam and the dashed lines indicate the electric circuit.

The pump light is modulated by a home-made electro-optic modulator (EOM) (crystal: MgO: $LiNbO_3$, dimension: 3 mm×3 mm×20 mm) with a modulation frequency of 14 MHz and a modulation index of $\beta = 0.4$ which are all fixed in the experiments reported in this paper. In order to avoid light feedback to perturb the laser, the frequency of the probe beam is shifted by 80 MHz using an acousto-optic modulator (AOM2). The probe beam is overlapped with the counter-propagating pump beam inside of an Yb HCL (Hamamatsu L2783-70NE-Yb), and both beam-diameters are about 1 mm. The HCL uses neon as the buffer gas and ytterbium as the cathode material with the main absorbed wavelength at 399 nm; its input and output windows are inclined at an angle of 10°. The probe beam and its sidebands which are generated by four-wave mixing among the probe and two components of the modulated pump are first reflected by PBS3 and then directed onto a fast photodiode (PD). The beat between the generated new sideband and the probe is amplified and de-modulated at the modulation frequency of 14 MHz using a double-balanced mixer (DBM). After passing through a low-pass amplifier (LP), the absorption spectrum of the ^{174}Yb on the transition $^1S_0$ to $^1P_1$ at 399 nm is observed and further used as the error signal for the frequency stabilization. This error signal is split into two parts, one is sent to a PID controller and then fed back to the piezoelectric transducer (PZT) of the ECDL as the slow feedback signal, the other is directly fed back to control the current of the ECDL as the fast feedback signal.

In order to perform frequency locking, we send a beam of light into an atomic beam apparatus as shown in Fig. 1. The atomic beam is produced by heating a sample of Yb inside a small oven with a temperature of about 623 K. At this temperature the vapour pressure of ytterbium is high enough to produce an atomic beam with sufficient intensity, emanating from the oven through a 3 mm small hole. After the atomic beam goes through a collimator with a diameter of 3 mm located at about 10 cm downstream from the oven, a highly collimated atomic beam is generated. On the other hand, the laser beam reflected by the PBS1 is first passed through an acousto-optic modulator (AOM3) having the centre frequency of 80 MHz and the bandwidth of 40 MHz. Then the first-order light output from AOM3 is collimated and sent into the vacuum chamber where the pressure during the experiment is lower than 1×10^{-6} Pa. The laser beam with the power of 0.1 mW and the diameter of 2 mm intersects with the ytterbium atomic beam at an angle of 90 ° inside of the chamber to minimize Doppler broadening and shifting of the laser frequency. Directly above the point where the atomic beam intersects with the laser light, a photomultiplier tube (PMT) is mounted. The fluorescence signal is detected by the PMT through a lens. Finally, the PMT output signal is sent to a computer for data analysis. Note that the windows of the vacuum compartment all are anti-reflection coated at 399 nm.

3. Experimental results

By optimizing the alignment of the pump beam with the probe beam inside of HCL shown in Fig. 1, we observed absorption spectra of all isotopes of ytterbium atoms except ^{168}Yb as shown in Fig. 2. The absorption signal of ^{174}Yb shown in the inset of Fig. 2 can be directly used as an error signal to lock the frequency of the laser and described by the expressions in Ref. [9], and its width is about 80 MHz, mainly determined by the pressure broadening effect which resulted from ytterbium atoms colliding with neon buffer gas in the HCL. Under typical operating conditions, the signal, benefiting from the use of modulation transfer spectroscopy technique, has large amplitude, steep slope and high sensitivity.

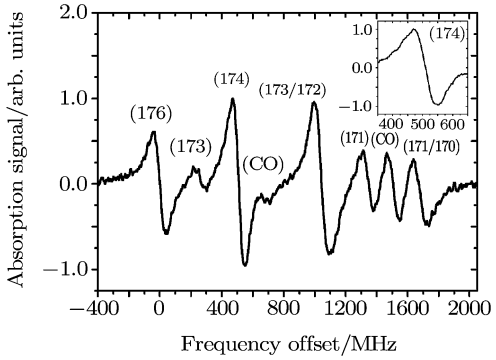


Fig. 2. Absorption spectra of ytterbium isotopes in the hollow cathode lamp on the transition at 399 nm by modulation transfer spectroscopy, where the probe power is 0.6 mW, pump power is 2.0 mW and HCL current is 4.0 mA. The inset is the absorption spectrum of ^{174}Yb. The CO stands for cross-over.

In order to better lock the laser, we need to maximize the magnitude and slope of the error signal. Therefore we have optimized parameters including the current of the HCL, powers of pump and probe beams for the absorption signal of ^{174}Yb as shown below.

Figure 3(a) shows the slope, magnitude and linewidth of the error signal dependences on the power of the pump beam. Note that, for simplicity, all signals have been normalized by their own maximum values. For the slope, there is a peak around 1.5 mW. In the low pump power region, the linewidth of absorption spectrum is weakly dependent on the pump power, and the amplitude of absorption signal rapidly increases with the increasing pump power, so that the slope is increased with the increasing pump power. Then in the high pump power region, the linewidth is increased with the increasing pump power due to the power broadening effect, and the magnitude goes to be constant, so that the slope is decreased with the increasing pump power. Considering there are two nearby optimum pump powers for the maximum slope and magnitude, respectively, we chose 2 mW as the optimum pump power for frequency locking. Figure 3(b) shows the slope, magnitude and linewidth of the error signal dependences on the probe beam power, which is similar to that in low pump power region as shown in Fig. 3(a). The slope is increased with the increasing probe power. In this case, the linewidth is almost independent of the probe beam power, and the amplitude of the signal is enhanced with the increasing probe power. Finally, the amplitude and slope of the signal are tapering off. In addition, since the maximum input power into the PD is limited to 1 mW, the optimum probe power was set to 0.6 mW for frequency locking.

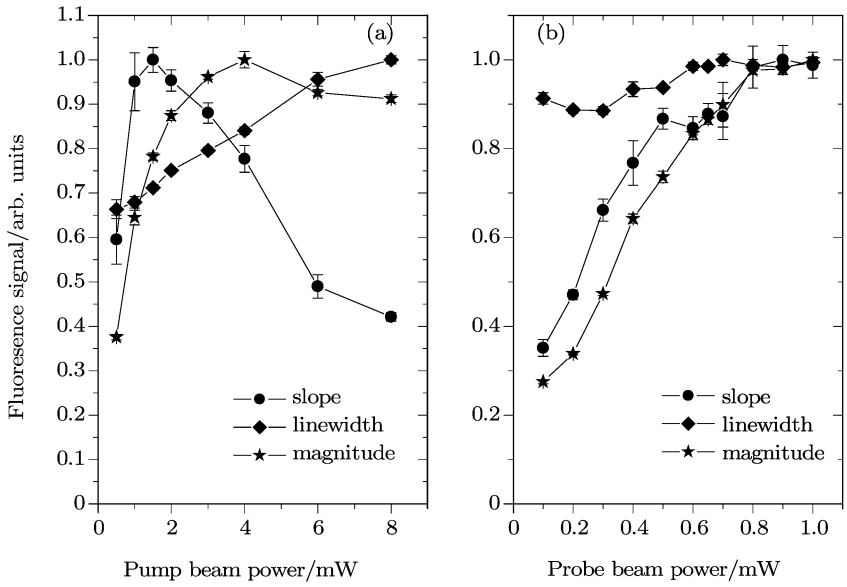


Fig. 3. Dependences of the slope at zero-crossing (circle), the magnitude (star), and the linewidth (diamond) of the error signal on (a) the pump beam power where the probe power is 0.6 mW, and (b) the probe beam power where the pump beam power is 2 mW, here the current of HCL is fixed at 3 mA.

Figure 4 shows the dependences of slope, magnitude and linewidth of the error signal on the current of the HCL. There are maxima for the slope and magnitude curves at a current of about 4 mA. We found that the linewidth decreased slowly with the increasing current. However, with the increasing current, the slope and magnitude increased in the low current region, but decreased in the high current region. A current of HCL at 4 mA was taken as the optimum value for frequency locking.

In order to have the optimum frequency locking signals, according to Figs. 3 and 4, the current of HCL and powers of pump and probe beams have been fixed at 4 mA, 2 mW and 0.6 mW, respectively. The typical signal-to-noise ratio of the optimum error signal was about 100 with a 100 Hz detection bandwidth as shown in the inset of Fig. 2. Using the optimum error signals, we have successfully locked the frequency of the 399 nm laser to the $^1S_0$ to $^1P_1$ transition of ytterbium atoms. Besides ^{174}Yb, we have also locked the laser frequency to the $^1S_0$ to $^1P_1$ transition of other Yb isotopes, for example, ^{176}Yb, ^{172}Yb, ^{171}Yb and ^{170}Yb respectively as shown in Fig. 2.

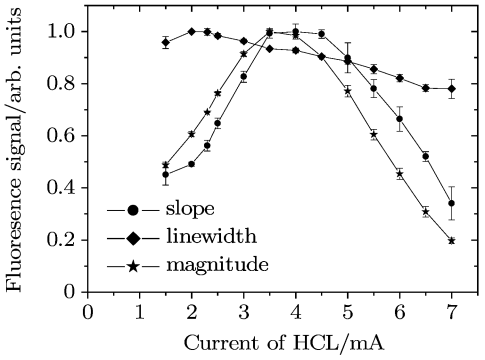


Fig. 4. Dependences of the slope at zero-crossing (circle), the magnitude (star), and the linewidth (diamond) of the error signal on the current of the HCL, where the pump and probe power is 2 mW and 0.6 mW respectively.

To demonstrate the frequency stabilization of the 399 nm laser, we measured the fluorescence signal fluctuation with time when the laser frequency was locked to the resonance frequency of the $^1S_0$ to $^1P_1$ transition of ^{174}Yb atoms as shown in Fig. 5.

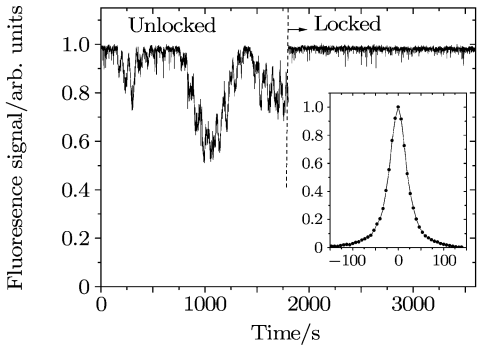


Fig. 5. Observing the resonance fluorescence signals in two cases. At first half of an hour: the laser is unlocked; at second half of an hour: the laser is locked. The inset shows the fluorescence spectrum of the ^{174}Yb atomic beam, when the input laser power is 0.1 mW, the oven temperature is 623 K and the PMT applied voltage is −350 V respectively.

The laser was interacted with the atomic beam coming from the oven with the temperature of 623 K as shown in Fig. 1. The inset of Fig. 5 shows the fluorescence spectrum profile of ^{174}Yb with the linewidth of about 40 MHz and the signal-to-noise ratio of 50 within a detection bandwidth of 1 kHz, therefore, the linewidth of the 399 nm laser could be estimated to be less than 1 MHz. By adjusting the frequencies of AOM1 and AOM3 to maximize the amplitude of the fluorescence signal, we easily found the laser frequency at resonance when 70 MHz is for AOM1 and 100 MHz for AOM3. For comparison, at first half of an hour, the laser was operating in an unlocked state, the fluorescence signal detected by the PMT which was applied with the voltage of −350 V randomly fluctuated with time as shown in the left part of Fig. 5; then at second half of an hour, the laser frequency is locked, the signal immediately went to be flat and stayed at the peak of the fluorescence signal as shown in the right part of Fig. 5, noting that there were some spikes mainly resulting from the stray scattering light entering the PMT detector spontaneously. Actually, the laser could be locked at this resonance frequency for a few hours and even longer.

Finally, we have quantitatively analysed the long-term frequency stability of the frequency-stabilized 399 nm laser by measuring the square-root Allan variance, $\sigma_y(\tau\mathrm{p})$, of the frequency fluctuation as a function of averaging time τ as shown in Fig. 6.

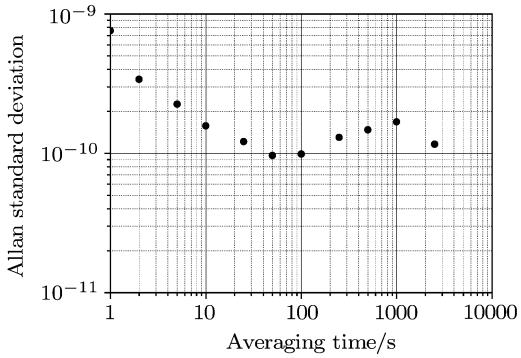


Fig. 6. Allan standard deviation of the 399 nm laser frequency as a function of averaging time.

In this measurement, we adjusted the frequency of AOM3 so that the frequency of the laser interacting with the Yb atomic beam was about half of the linewidth away from the resonance frequency, and the intensity of the fluorescence signal was mainly sensitive to the frequency fluctuation of the laser. The frequency stability of the 399 nm laser was calculated from the Allan variance. From Fig. 6, we could find the long-term frequency stability of the stabilized 399 nm laser was less than 2×10^{-10} (corresponding to 300 kHz) for 2500-s averaging time, noting that the true frequency stability should be better than the value we presented here because the other fluctuations were not ruled out from our data, for instance, the fluctuations of the laser power, laser direction and Yb atomic beam flux, etc.. Actually, to measure the frequency stability precisely, one should do the beat frequency measurement between two identical lasers. Unfortunately, we do not have the second 399 nm laser at this moment. In addition, we also observed that the frequency drift of the stabilized 399 nm laser was less than 1 MHz within a few hours. These results show that the frequency-stabilized 399 nm laser described in

this paper by using the MTS technique is good enough to be employed in the experiments on the laser cooling and trapping of Yb atoms.

4. Conclusion

In summary, we have successfully locked the frequency of the 399 nm laser to the $^1S_0$ to $^1P_1$ transition of ytterbium atoms in a hollow cathode lamp by using the MTS technique under the optimum conditions. We could further improve the error signals by optimizing the parameters of the EOM, including the modulation index and frequency.[9] The scheme presented in this paper is more effective and simple, and the apparatus is more compact and flexible. This method can be easily applied to other laser systems with different types of atomic species. Currently, the stabilized 399 nm laser has been used to explore experiments on the first-stage cooling and trapping of ytterbium atoms, and the cold atoms have been produced efficiently and the stable Yb $^1S_0$ to $^1P_1$ MOT can be maintained for at least eight hours. In addition, we will use this stabilized 399 nm laser to build a precision spectrometer to measure the isotope shifts of all ytterbium isotopes precisely.

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