A linewidth-narrowed and frequency-stabilized dye laser for application in laser cooling of molecules

D. P. Dai, Y. Xia,[*] Y. N. Yin, X. X. Yang, Y. F. Fang, X. J. Li, and J. P. Yin

State Key Laboratory of Precision Spectroscopy, Department of Physics, East China Normal University, Shanghai 200062, China

[*]*yxia@phy.ecnu.edu.cn*

Abstract: We demonstrate a robust and versatile solution for locking the continuous-wave dye laser for applications in laser cooling of molecules which need linewidth-narrowed and frequency-stabilized lasers. The dye laser is first stabilized with respect to a reference cavity by Pound-Drever-Hall (PDH) technique which results in a single frequency with the linewidth 200 kHz and short-term stabilization, by stabilizing the length of the reference cavity to a stabilized helium-neon laser we simultaneously transfer the ± 2 MHz absolute frequency stability of the helium-neon laser to the dye laser with long-term stabilization. This allows the dye laser to be frequency chirped with the maximum 60 GHz scan range while its frequency remains locked. It also offers the advantages of locking at arbitrary dye laser frequencies, having a larger locking capture range and frequency scanning range to be implemented via software. This laser has been developed for the purpose of laser cooling a molecular magnesium fluoride beam.

OCIS codes: (020.3320) Laser cooling; (140.3425) Laser stabilization; (140.3600) Lasers, tunable.

References and links

1. M. D. Di Rosa, "Laser-cooling molecules," Eur. Phys. J. D **31**(2), 395–402 (2004).
2. B. K. Stuhl, B. C. Sawyer, D. Wang, and J. Ye, "Magneto-optical trap for polar molecules," Phys. Rev. Lett. **101**(24), 243002 (2008).
3. E. S. Shuman, J. F. Barry, and D. DeMille, "Laser cooling of a diatomic molecule," Nature **467**(7317), 820–823 (2010).
4. M. T. Hummon, M. Yeo, B. K. Stuhl, A. L. Collopy, Y. Xia, and J. Ye, "2D Magneto-optical trapping of diatomic molecules," Phys. Rev. Lett. **110**(14), 143001 (2013).
5. V. Zhelyazkova, A. Cournol, T. E. Wall, A. Matsushima, J. J. Hudson, E. A. Hinds, M. R. Tarbutt, and B. E. Sauer, "Laser cooling and slowing of CaF molecules," Phys. Rev. A **89**(5), 053416 (2014).
6. E. L. Raab, M. Prentiss, A. Cable, S. Chu, and D. E. Pritchard, "Trapping of neutral sodium atoms with radiation pressure," Phys. Rev. Lett. **59**(23), 2631–2634 (1987).
7. J. Doyle, B. Friedrich, R. V. Krems, and F. Masnou-Seeuws, "Ultracold polar molecules: formation and collisions," Eur. Phys. J. D **31**(2), 149–445 (2004).
8. L. D. Carr, D. DeMille, R. V. Krems, and J. Ye, "Cold and ultracold molecules: science, technology and applications," New J. Phys. **11**(5), 055049 (2009).
9. R. Krems, B. Friedrich, and W. C. Stwalley, *Cold Molecules: Theory, Experiment, Applications* (CRC, 2009).
10. D. S. Jin and J. Ye, eds., Chemical Reviews, Special Issue on Ultracold Molecules **112**, 4801–5072 (2012).
11. E. S. Shuman, J. F. Barry, D. R. Glenn, and D. DeMille, "Radiative force from optical cycling on a diatomic molecule," Phys. Rev. Lett. **103**(22), 223001 (2009).
12. J. F. Barry, D. J. McCarron, E. B. Norrgard, M. H. Steinecker, and D. DeMille, "Magneto-optical trapping of a diatomic molecule," Nature **512**(7514), 286–289 (2014).
13. F. J. Duarte, L. W. Hillman, P. F. Liao, and P. Kelley, *Dye Laser Principles with Applications* (Academic, 1990), Chap. 5.
14. J. Helmcke, S. A. Lee, and J. L. Hall, "Dye laser spectrometer for ultrahigh spectral resolution: design and performance," Appl. Opt. **21**(9), 1686–1694 (1982).

15. R. W. P. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward, "Laser phase and frequency stabilization using an optical resonator," Appl. Phys. B **31**(2), 97–105 (1983).
16. J. Hough, D. Hils, M. D. Rayman, L. S. Ma, L. Hollberg, and J. L. Hall, "Dye-laser frequency stabilization using optical resonators," Appl. Phys. B **33**(3), 179–185 (1984).
17. M. Zhu and J. L. Hall, "Stabilization of optical phase/frequency of a laser system: application to a commercial dye laser with an external stabilizer," J. Opt. Soc. Am. B **10**(5), 802–816 (1993).
18. B. G. Lindsay, K. A. Smith, and F. B. Dunning, "Control of long-term output frequency drift in commercial dye lasers," Rev. Sci. Instrum. **62**(6), 1656–1657 (1991).
19. E. Riedle, S. H. Ashworth, J. T. Farrell, Jr., and D. J. Nesbitt, "Stabilization and precise calibration of a continuous-wave difference frequency spectrometer by use of a simple transfer cavity," Rev. Sci. Instrum. **65**(1), 42–48 (1994).
20. J. Biesheuvel, D. W. E. Noom, E. J. Salumbides, K. T. Sheridan, W. Ubachs, and J. C. J. Koelemeij, "Widely tunable laser frequency offset lock with 30 GHz range and 5 THz offset," Opt. Express **21**(12), 14008–14016 (2013).
21. C. W. Oates, K. R. Vogel, and J. L. Hall, "High precision linewidth measurement of laser-cooled atoms: Resolution of the Na 3p $^2P_{3/2}$ lifetime discrepancy," Phys. Rev. Lett. **76**(16), 2866–2869 (1996).
22. V. Wippel, C. Binder, W. Huber, L. Windholz, M. Allegrini, F. Fuso, and E. Arimondo, "Photoionization cross-sections of the first excited states of sodium and lithium in a magneto-optical trap," Eur. Phys. J. D **17**(3), 285–291 (2001).
23. T. W. Hansch and B. Couillaud, "Laser frequency stabilization by polarization spectroscopy of a reflecting reference cavity," Opt. Commun. **35**(3), 441–444 (1980).
24. J. L. Hall and T. W. Hänsch, "External dye-laser frequency stabilizer," Opt. Lett. **9**(11), 502–504 (1984).
25. MgF molecule has a main cooling transition on $X^2S^+ \rightarrow A^2P_{1/2}$ at 359.3nm, highly diagonal Franck-Condon factors limit the vibrational branching of $A^2P_{1/2}$, only two additional lasers at 368.6nm and 368.1nm to repump the v" = 1, 2 levels are needed to limit the vibrational branching loss to $<10^{-6}$. These lasers are from dye laser frequency doubling in a doubling cavity.
26. The linewidth of laser frequency is derived from the relative frequency deviation (RMS deviation) from the current lock frequency of the reference cell calculated with the help of the PDH error function for the resonator with a free spectral range and a finesse, which is analyzed by the digital signal processor software of Matisse dye laser.

1. Introduction

In the past few years, a new approach, laser cooling and trapping of diatomic molecules has become possible [1–5]. The development of a molecular MOT should really mirror the huge historical success achieved by the atomic MOT [6]. Realizing such a powerful technique for producing a diverse set of dense, ultracold diatomic molecular species will open a new chapter for molecular science and it will greatly advance understandings in precision measurement, strongly correlated many-body quantum systems and physical chemistry in the most fundamental way [7–10]. Until now, radiative force from optical cycling [11], Doppler cooling [3] and the 2D [4], 3D [12] magneto-optical trap have been demonstrated using the $X^2S^+ \rightarrow A^2P_{1/2}$ electronic transition of laser-cooling diatomic molecules (SrF [3], YO [4], CaF [5]). Due to the large number of diatomic molecules, there are many more candidates as well suitable for laser cooling experiment. The laser wavelengths of a quasicycling transition of molecules are covered from ultraviolet to infrared radiation. The wavelengths for less of candidate molecules are accessible by diode lasers [2,3], most of them need continuous-wave (cw) dye laser or Titanium: Sapphire laser to provide the light source for cooling and trapping experiments [4,5].

As we know, stabilizing the cw dye laser is important for some high-resolution spectroscopy applications and laser cooling experiments. Most of dye laser locking methods can be applied equally well to stabilize the frequency of lasers to a cavity or to atomic or molecular resonant spectra line [13–24]. There are several kinds of locking techniques for dye laser frequency stabilization. Cavity-side-locking method uses the difference between the photoncurrents of the spectrally sharp cavity transmittance peak and the laser power as the error signal to lock the dye laser to the side of the cavity [14]. The other dye laser stabilization method has developed using RF heterodyne techniques (PDH technique), which has both center frequency short-term stabilization and the narrowest linewidths [15–17]. Another method is that using a scanning confocal FP etalon and a stabilized HeNe laser controls long-term frequency drift of the ring dye laser to several MHz stability, but laser frequency linewidth doesn't be narrowed [18–20]. The dye laser is also stabilized with an external FP cavity and locked to atomic or molecular transition line, offered a narrowed linewidth and long-term frequency stability, but it restricts the continuous tuning capability and arbitrary dye laser frequencies locking when locking on transition line [21,22]. In addition, polarization locking method [23], locking scheme combining an acousto-optic frequency shifter with a fast electro-optic phase modulator [24], and optical heterodyne stabilization from a self-referenced octave-spanning erbium-doped fiber frequency comb [4] are also performed.

For the present experiments of laser-cooling molecules, the pulsed molecular beam is produced by pulsed YAG laser ablation from solid targets [3–5], which cannot provide stable saturated absorption spectroscopy to stabilize the cooling and repumping laser frequency. Whereas the frequency stabilization of cw dye laser using atomic or molecular spectra line as reference only can work at some special wavelengths, the atomic and molecular resonances cannot provide the same signal-to-noise ratio that is available from a cavity, but their long-term stability is generally much better. For dye lasers to have high resolution and high accuracy, it is customary to lock the laser's frequency to a high-finesse reference cavity (to achieve the narrow linewidths) and then to lock the cavity to an atomic or molecular resonance (which then provides the long-term stability) [13]. Here we demonstrate a frequency stabilization scheme to prepare the linewidth-narrowed and frequency-stabilized lasers with a larger scanning range for laser cooling of molecules. To stabilize the dye laser frequency, two different locking techniques are used in our experiment. The operating scheme for locking dye laser using two cavities and to stabilities of about ± 2 MHz is a fundamental component of an ongoing experiment of laser-cooling magnesium fluoride (MgF) [25]. If there isn't an active control on the laser frequency, the ring cavity of the dye laser is not sufficiently stable for staying on resonance with the molecular transition for long periods of time, the laser frequency drifts by more than 600 MHz/hour, if we lock this laser on the reference cavity by PDH locking, the frequency deviation is about 80 MHz/hour due to the ambient temperature change and piezo actuator relaxation. To get rid of long-term drifts, we can preserve the length of the PDH reference cavity a constant. We use a method of scanning cavity to transfer the frequency stabilization of HeNe laser to the single-mode dye laser, and the feedback is sent to the piezo actuator of the PDH reference cavity. All the controlling system of the laser stabilization is realized using software program. Then we will use frequency doubled second harmonic of the stabilized dye laser to demonstrate the quasi-cycling transition in MgF molecule.

2. Experimental setup

The experimental setup is shown in Fig. 1. The commercial dye laser (Matisse DX) consists of pump laser, dye laser head and PDH reference cavity. The single-mode selection is realized by different frequency selective elements. The ring resonator length of the dye laser is 1.7 m with mode spacing ~165 MHz. Birefringent Filter as coarse wavelength selection has free spectral range (FSR) of ~100 nm, the Thin Etalon, FSR ~250 GHz, and the Piezo Etalon, FSR ~19 GHz, and the large number of longitudinal modes of the cavity is within the bandwidth of the etalon, so the mode competition between the ring cavity and homogeneous broadened medium generates the sing-mode operation.

With 6 W 532 nm of pump light, the dye cavity well optimized we have achieved output powers of up to 1.2 W at a wavelength of 577 nm as the testing light. Without active control the laser linewidth is broadened to about 20 MHz, and the free-running frequency deviation is more than 600 MHz/hour. In order to stabilize the dye laser frequency a fraction of the laser is directed into a vacuum-insulated and temperature-controlled reference cavity, the temperature resolution is 0.1 K, it is the PDH reference cavity part in Fig. 1. The laser beam passes through an electro-optical modulator which adds sidebands to the original beam. The sidebands are off-resonant, the original beam is resonant (EOM 20 MHz shift; 5.2 MHz cavity linewidth). The original beam will acquire phase information from the cavity. All beams are

#222958 - $15.00 USD Received 11 Sep 2014; revised 23 Oct 2014; accepted 5 Nov 2014; published 10 Nov 2014

superimposed on a photodetector and mixed. An RF-mixer extracts the part of the signal that varies with the modulation frequency. The phase-shifter balances the signal to be zero for the case that the original laser is resonant. The error signal is filtered and amplified to drive a servo. The servo pulls the laser emission frequency to be resonant with the cavity. The fast piezo(M3) and slow piezo(TM) in the ring-cavity of the dye laser will automatically adjust the ring cavity length according to the feedback PDH error signal when the dye laser is locked to the reference cavity, which has FSR of 1300 MHz and finess of 250. The linewidth narrowing down to a magnitude of one hundred kHz is achieved by introduction of an intra-cavity electro-optical modulator into the dye ring laser. The dye laser is stabilized with respect to this reference cavity which results in a single frequency laser linewidth of about 200 kHz [26]. But its frequency will drift as the length of the reference cavity changes, which is caused by the temperature fluctuation and the piezo actuator relaxation. The PDH locking is benefit for the narrowing of laser linewidth and short-term stabilization of dye laser frequency, and it has important advantages over other locking schemes, such as wide locking range, insensitive to intensity fluctuations, high sensitivity to frequency fluctuations. The PDH locking are realized and optimized by the digital signal processor software of the dye laser.

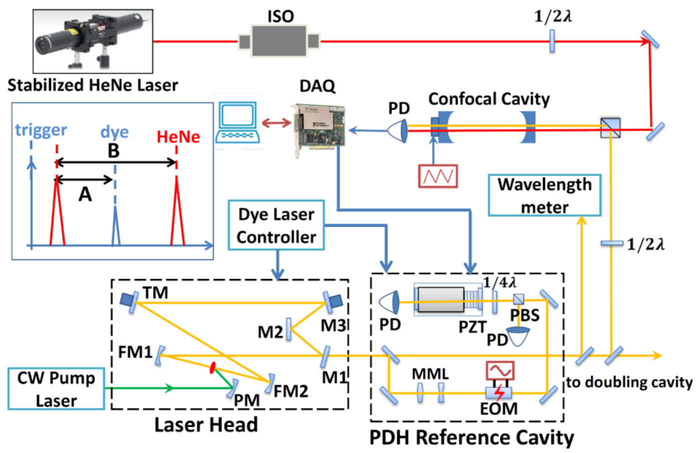

Fig. 1. Experimental layout. CW pump laser, dye laser head and PDH reference cavity; Stabilized HeNe laser; ISO optical isolator; PD photodetector; EOM Electro-optical Modulator; λ/2 half waveplate; MML Mode matching lens; λ/4 quarter waveplate; PBS Polarization beam splitter; DAQ Data Acquisition Card. The confocal cavity works as a transfer cavity. The inset shows the transmission peaks of stabilized HeNe laser and dye laser.

In order to stabilize the dye laser against such frequency fluctuations (long-term stability) we compare its frequency to the frequency of a stabilized HeNe laser via a transfer cavity. A frequency-stabilized HeNe (Thorlabs HRS015) with a frequency stability ~ ± 2 MHz in eight hours is used to stabilize cw dye laser frequency to ~ ± 2 MHz. Dye laser is locked to a PDH reference cavity, in turn, which is locked to the HeNe laser. The stabilized HeNe laser beam passes through an optical isolator and is transmitted through the beamsplitter into a confocal cavity (Toptica FPI100) with FSR of 1.0 GHz and finess of 400. The length of the transfer cavity is scanned by applying a voltage sawtooth (0, 10 V) and a variable DC offset on a piezoelectric transducer which is attached to one mirror. This voltage ramp is supplied by a function generator at a rate of 50 Hz. When the cavity is on resonance with HeNe laser the light is transmitted through the cavity, it is detected by the photodiode. A small fraction of the dye laser beam is sent through the same transfer cavity onto the same photodiode. The voltage signal from the photodiode is then read into the computer via a data acquisition card. The setup of the transfer cavity locking is shown in Fig. 1. It consists of a function generator, a frequency-stabilized HeNe laser at 632.9918 nm, a Topical confocal cavity and the data

#222958 - $15.00 USD Received 11 Sep 2014; revised 23 Oct 2014; accepted 5 Nov 2014; published 10 Nov 2014

acquisition system, in addition, a cw dye laser with PDH locking. A photo detector is integrated in the end of the confocal cavity, a tunable resistant is used to transfer the photo current signal into voltage signal.

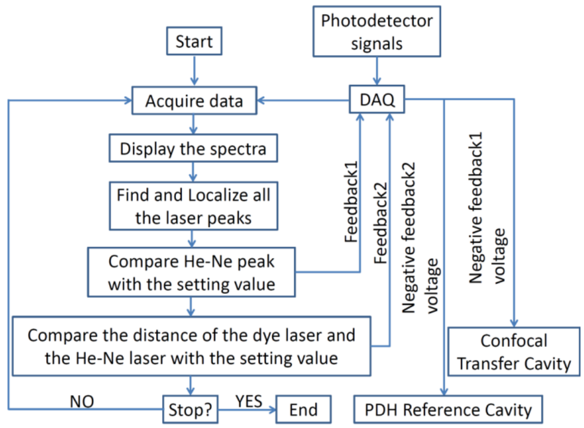

Fig. 2. The flow chart of the laser stabilization experiment.

Triangular wave provided by functional generator is used to scan the length of the transfer cavity. The rate of the scanning is 50 Hz, and the peak-to-peak amplitude of the triangular wave is 10 volts to promise one FSR shown. The inset of Fig. 1 shows the transmission peaks of the lasers. The transmission-power data are digitalized using a DAQ card (NI PCI-6259), the digital data is then sent to the Labview program designed by ourselves. This program will calculate one feedback voltage signal to control the length of the piezo of the PDH reference cavity.

The flow chart is shown in the Fig. 2 and the concrete operation of the experiment is done as the following. In the half of the triangular wave, the DAQ card acquires the transmission-power information. The computer algorithm will find out the positions of the transmission peaks of the two lasers. Although the HeNe laser is frequency stabilized, the position of the transmission peak will still move due to the temperature fluctuations and piezo actuator relaxation. So the first step is to lock the position of the HeNe transmission peak according to the difference between the position of the HeNe transmission peak and the locking point. A specific voltage will be decided using PI algorithm to add on the triangular wave as an offset voltage, so the position of HeNe transmission peak is locked and the influence of the temperature fluctuation is canceled out successfully. Then we should lock the position of the transmission peak of the dye laser to the transmission peak of HeNe laser. We set the input voltage produced from an analog output channel of the DAQ card, through amplification, to add on the piezo actuator of the reference cavity gradually, which has a slow rate of 5 mV/s. The rate is so slow to promise the dye laser is always locked onto the reference cavity which is realized by PDH technique. The computer will calculate the difference between the position of the transmission peak of the dye laser and the position of the first transmission peak of the HeNe laser, denoted A here. The distance between two consecutive transmission peaks of He-Ne laser can also be acquired, denoted B here. We choose a constant C between 0 and 1 as our locking point, and the difference between A/B and C is referred as the error signal, according to which another PI algorithm is designed to lock the difference between A/B and C to zero. From the error signal Labview program can detect which direction the frequency has drifted to and compensate the frequency drift by changing the voltage of the PDH reference cavity. For the confocal cavity, as we know, the signal generator is used to repetitively scan the length of the confocal cavity by $\lambda/4$ in order to sweep through one FSR of the interferometer. From the inset of the Fig. 1 we can

#222958 - $15.00 USD Received 11 Sep 2014; revised 23 Oct 2014; accepted 5 Nov 2014; published 10 Nov 2014

get $N_M \times \lambda_M/(4n_M) + A/B \times \lambda_M/(4n_M) = N_S \times \lambda_S/(4n_S)$, from which $\lambda_S = n_S/n_M \times \lambda_M \times (N_M + \alpha)/N_S$ can be obtained, where $\alpha = A/B$, λ_M, λ_S are the wavelength of the HeNe laser and dye laser, and N_M, N_S are the resonance peak modes of the two lasers, n_M, n_S are the refractive index in air for HeNe laser and dye laser, respectively. This equation can be used to determine the wavelength of the dye laser according to the position of the transmission peak of the dye laser. The resonance peak modes N_M and N_S will not change as long as the reference cavity is locked to the HeNe laser, although the ambient temperature is changing as the time.

3. Results

We set the frequency of the triangular wave exerted on the transfer cavity as 50 Hz. The sampling rate of DAQ card is set as 1 MS/s, so we can acquire 10000 data points during half of the period of the triangular wave. The software will analyze these data points and get the negative feedback voltage of the transfer cavity and the reference cavity to lock the localizations of these peaks.

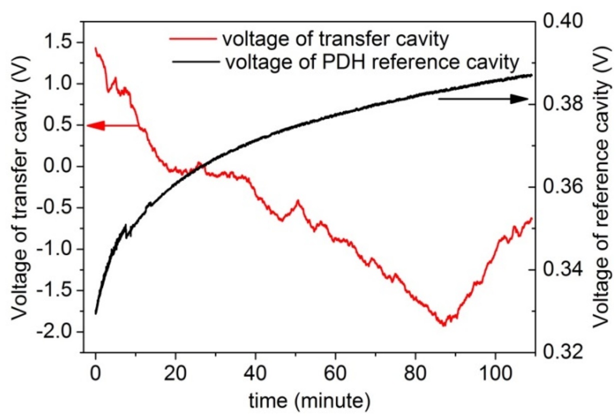

Fig. 3. The voltage of the transfer cavity and the reference cavity via time when the frequency of the triangular wave is 50 Hz during the time of 100 minutes. Both voltages are ones before amplification.

Figure 3 gives how the voltage of the transfer cavity and reference cavity change via time during 100 minutes when the reference cavity is locked onto the HeNe laser via the transfer cavity. From the figure we can find that the voltage of the transfer cavity is changing to compensate the fluctuation of the temperature and the flow of the air, and the voltage is fluctuating in the short term because the ambient temperature and air flow are fluctuating and random. The voltage of the transfer cavity is decreasing for about 85 minutes before increasing again, because the ambient temperature is changing in the opposite direction at about the 85th minute. The voltage of the PDH reference cavity is becoming larger, this is because the length of the reference cavity is drifting in one direction during the time of 100 minutes. But the rate of the increase of this voltage is becoming slower, the reason is that a specific voltage is added onto the piezo actuator of the reference cavity at a very slow rate before we start locking the reference cavity onto the HeNe laser via transfer cavity, the system is not that stable at the beginning and then becoming more and more stable, so the rate of the change of this voltage is becoming slower and the voltage of the reference cavity will oscillate at a slow rate in the future.

#222958 - $15.00 USD Received 11 Sep 2014; revised 23 Oct 2014; accepted 5 Nov 2014; published 10 Nov 2014

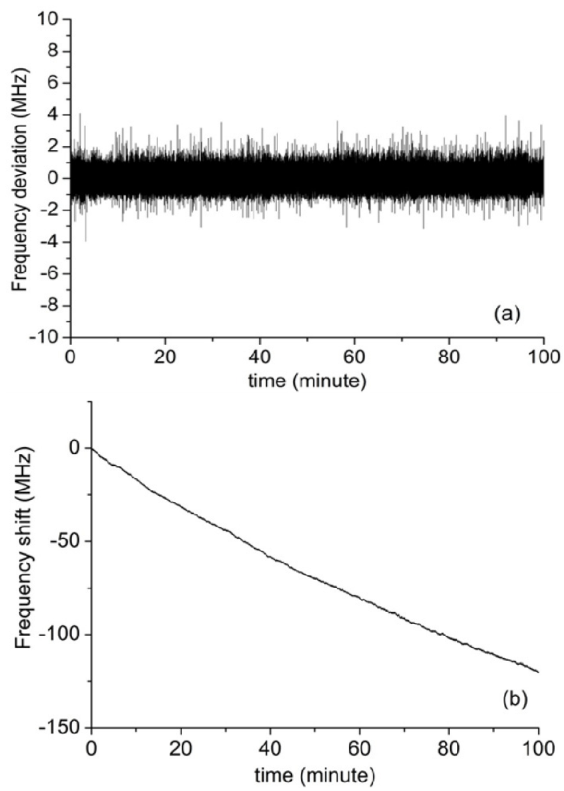

Fig. 4. (a) Frequency deviation of dye laser relative to HeNe laser when it is locked using scanning cavity; (b) frequency drift of the dye laser when it is only locked on PDH reference cavity, the drift value is relative to the initial frequency at 519.194283 THz.

The accuracy of the dye laser frequency relative to HeNe laser is calculated using the formula $(\mathrm{A}/\mathrm{B}-\mathrm{C})\times\mathrm{FSR}\times\lambda_{\mathrm{M}}/\lambda_{\mathrm{S}}$. The precision of the frequency of the dye laser is shown in Fig. 4(a), from which we can get we have stabilized the frequency of the dye laser to $\approx\pm 2\mathrm{MHz}$ precision. This has met the requirement for the stability of laser frequency in the experiment of laser cooling of MgF molecules. Figure 4(b) gives the frequency drift of dye laser when it is only locked on the PDH reference cavity, from the figure we know the dye laser can drift hundreds of MHz range per 100 minutes, which proves our method of frequency stabilization has worked well.

Table 1 shows the summary of the locking parameters for dye laser when it is free-running without PDH locking and transfer cavity, is with PDH locking, and is with PDH locking and transfer cavity. It is shown from Table 1 that PDH locking narrows the linewidth of dye laser to 0.20 MHz, the short-term frequency drift is effectively suppressed, and transfer cavity locking corrects and controls the long-term frequency deviation, combination with both locking methods works well for frequency deviation and linewidth narrowing of dye laser.

Table 1. Locking Parameters of Dye Laser under Free-running, PDH Locking, and Transfer Cavity Modes

Parameters of dye laser	Free-running	With PDH locking	With PDH locking and transfer cavity
Frequency deviation for a long-term period	> 600 MHz/hour	~80 MHz/hour	< ± 2 MHz/hour
Linewidth of dye laser	20 MHz	~0.20 MHz [26]	~0.20 MHz [26]

#222958 - $15.00 USD Received 11 Sep 2014; revised 23 Oct 2014; accepted 5 Nov 2014; published 10 Nov 2014

4. Conclusion

In summary, we have reported long-term frequency stabilization and linewidth narrowing of the cw dye laser for application in laser cooling of molecules, which is realized by combining Pound-Drever-Hall locking with scanning transfer cavity technique. Using this technique we can lock the dye frequency to the stabilized HeNe reference with a ± 2 MHz stability, the software further allows to accurately scan the laser within the maximum 60 GHz of cavity and to apply very precise frequency detuning within 1.0 GHz FSR of transfer cavity. This kind of stabilization system is particularly useful when no atomic or molecular reference lines are available, as in the case of many stabilized lasers needed which is via one transfer cavity.

Acknowledgments

We acknowledge support from the National Natural Science Foundation of China under grants 11374100, 11034002, the Natural Science Foundation of Shanghai Municipality (Grant No. 13ZR1412800), the National Key Basic Research and Development Program of China under Grant Nos. 2011CB921602.