

Narrow-linewidth light source for a coherent Raman transfer of ultracold molecules

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Abstract: We describe a tunable two-color CW light source sufficient for realizing a coherent Raman transfer between two molecular states that are more than 0.5 eV (120 THz) apart. The simultaneous frequency stabilization of 901 nm and 655 nm light was achieved by locking diode lasers to a single ultralow expansion cavity with dual wavelengths coating. By utilizing offset-locking and optical phase-locked loop (OPLL), we ensured a large mode-hop free tuning range (> 2 GHz). The obtained short term linewidth (< 10 Hz) and the linear drift of frequency (65 mHz/s) were both sufficient to eliminate the influence of laser linewidths on the efficiency of coherent Raman transition.

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Narrow-linewidth lasers have comprised a crucial part of a wide variety of experiments in atomic, molecular, and optical physics, including frequency standards [1,2] and atomic spectroscopy [3]. Recent advances in manipulating ultracold atoms have opened a new application of narrow-linewidth lasers in producing ultracold molecules in the lowest rovibrational level from associated pairs of ultracold atoms via stimulated Raman adiabatic passage (STIRAP) [4–6]. In these experiments, a narrow-linewidth two-color light source with a large frequency difference plays a key role. The efficiency of Raman transfer is determined by the coherence between the two frequencies. Hence, it is necessary to develop a general method for producing a mutually coherent, two-color light source with a large frequency difference corresponding to the binding energy of a molecule (a few hundred THz for alkali diatomic molecules). In previous studies, two frequencies were stabilized independently by locking lasers directly to a frequency comb [4], or to multiple cavities referenced to a single frequency comb [5]. In this letter, we show that a simple laser system based on a single ultralow expansion (ULE) cavity with dual-wavelength coating provides both tunability and coherence required for an efficient STIRAP transfer. Narrow linewidth is achieved by optimizing electronic and optical feedback to master lasers, while long term stability is ensured by operating ULE cavity at zero-expansion temperature. Large mode-hop free tuning range needed for molecular spectroscopy (over a few GHz) is guaranteed by utilizing optical phase-lock loop to slave lasers. The influence of vibration on the cavity, which is the only remaining unknown factor, is estimated by measuring the acceleration on the table. Our results provide a general method for achieving two narrow-linewidth lasers to a single cavity with moderately high-finesses (6000–10000).

Figure 1 and 2 shows the entire optical setup. The ULE cavity is kept in a vacuum chamber, which is mounted on a passively stabilized optical table (Minus-K, 250BM-1). Since our vacuum chamber was based on the design given in Ref [3], we briefly describe our chamber herein. In our experiments, a temperature stability of 0.1 K near the zero-expansion temperature is sufficient because the typical two-photon linewidth of our STIRAP experiments is on the order of 100kHz. We achieved this stability by surrounding the ULE cavity with two nested heat shields and stabilizing the outer shield with a double stage Peltier element. The zero-expansion temperature of our ULE cavity was determined to be 6.6 °C by monitoring the transmission line referenced to the Cs D2 line. The long-term stability was checked by measurements on the narrow transition of $^{41}\text{K}^{87}\text{Rb}$ molecule ($X^1\Sigma^+-(3)^1\Sigma^+$; natural linewidth, 320(30) kHz), which showed a linear drift with a slope of 65(1) mHz/s (Fig. 3). This drift arises from the aging of the ULE glass and is comparable to previous study [3]. Since this drift is linear over months, it is readily compensated by subtracting it as an expected offset. Thus, we obtain a long-term stability of better than 100 kHz as shown in Fig. 3.

Vibration isolation is crucial since it directly influences the STIRAP efficiency. We employed a notched horizontal cavity because a notched cavity has a lower sensitivity to vibration [7]. The cavity was supported at calculated Airy points, and the Minus-K optical table was surrounded by sound insulation boards including a lead sheet with a width of 0.5 mm (Toho Zinc, P-2) for minimizing the influence of acoustic noise. We measured the acceleration of the optical table by using a seismic accelerometer (Wilcoxon, 731A/P31; sensitivity, 1 kV/g; noise, 0.03 $\mu\text{g Hz}^{-1/2}$ at 2 Hz, where g is acceleration of gravity). Figure 4 shows the measured acceleration. Assuming the typical sensitivity of a notched cavity (< 1 MHz/g in a horizontal direction and < 100 kHz/g in a vertical direction [7]), we concluded that our cavity should be able to provide the frequency stability better than 10 Hz.

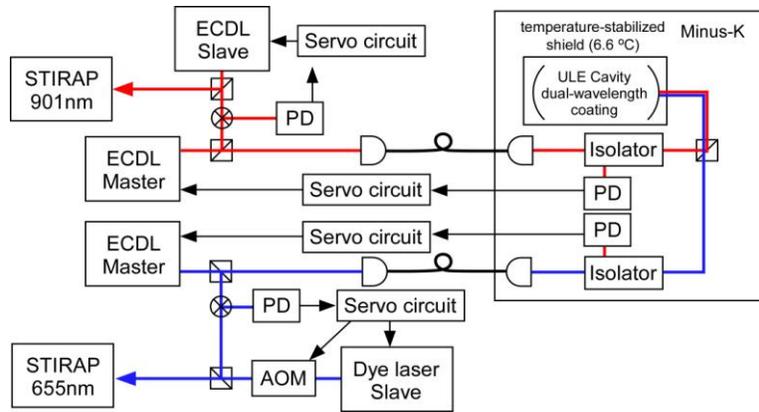


Fig. 1. Optical setup of the two-color light source. Two external cavity diode lasers (ECDL) work as master lasers which are directly locked to the ULE cavity with dual-wavelength coating. The light of the master lasers is carried through polarization maintaining fibers (10 m). The linewidth of the master lasers is narrower than 10 Hz and its long-term stability is better than 100 kHz for a few days. STIRAP experiments are carried out with two slave lasers which are offset-locked to the master lasers via OPLL. The slave lasers are also used for the beat note measurement for measuring the linewidth of the master lasers, when the slave lasers are locked to the ULE cavity.

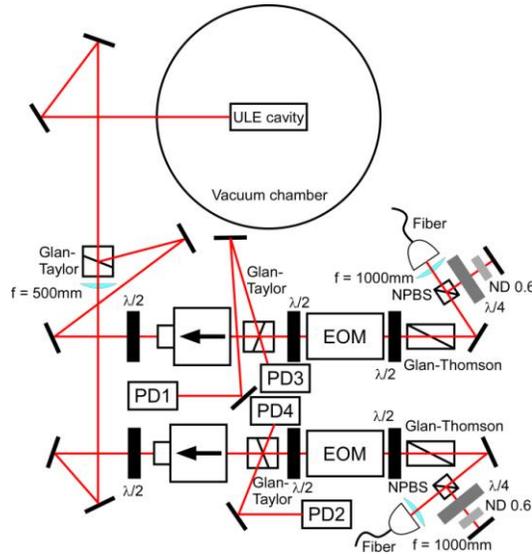


Fig. 2. Actual optical setup around the ULE cavity. The light from a master laser is carried by an optical fiber to the table. A part of the light is back-reflected to the fibers in order to cancel the phase noise in the fibers. The Glan-Thomson polarizer is used to purify the polarization for minimizing the variation in the offset of the error signal. The half waveplates before and after the EOM are used to cancel the offset of the error signal. The isolator has two roles. One is to extract the reflected light from the cavity for obtaining the error signal. The other is to prevent the reflected light from going back to the fiber. The two light is finally overlapped with a Glan-Taylor polarizer and is incident on the cavity. The typical power incident on the cavity is 200 μ W for both lights. In order to avoid interferences between the cavity and any other optics, all the optics including isolators are tilted against the optical axis. The PD1 and PD2 are used for the PDH locking, whereas the PD3 and PD4 are used for stabilizing the laser power to within 0.2%.

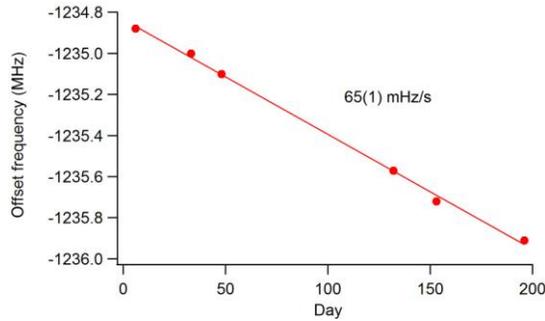


Fig. 3. Long-term stability of the ULE cavity measured with a molecular transition. The offset frequency between a laser locked to the ULE cavity and a laser monitoring molecules is plotted with respect to date. The slope of 65 mHz/s is comparable to the previous study [3] and is understood as the aging of the ULE glass. The observed linear drift indicates that the fluctuation due to the variation in room temperature is well below 100 kHz.

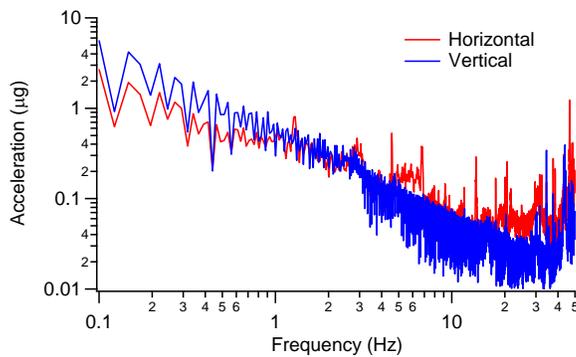


Fig. 4. Acceleration of the Minus-K optical table used for mounting the ULE cavity. The acceleration at 1 Hz is well below 10 μg . Since a notched cavity suspended at Airy points has a typical sensitivity lower than 100 kHz/g in a horizontal direction and 10 kHz/g in a vertical direction, our cavity is expected to provide a frequency stability better than 10 Hz for a few seconds.

In our setup, frequency stability is determined by master lasers. Thus, the narrow linewidth of the master laser is one of the crucial requirements for STIRAP experiments. Here we briefly describe our setup for achieving the narrow linewidth. Our master laser was a standard grating-stabilized external cavity diode laser (ECDL) [8] locked to the ULE cavity via the Pound-Drever-Hall (PDH) method [9]. For 901 nm, we used an antireflection (AR) coated laser diode (Eagleyard, EYP-RWE-940) which covered 870-920 nm. For 655 nm, we used a non-AR-coated laser diode (Roithner lasertechnik, ADL-65401TU) which could cover several nm.

The linewidth of the ECDL without any feedback was approximately 100 kHz. For narrowing the linewidth, both current and piezo feedbacks were used. The finesse of our cavity was measured as 6000 at 901 nm and 10000 at 655 nm by using the dynamic response of a Fabry-Perot cavity [10]. The current feedback had a role of suppressing pedestals at high frequencies and making the lock robust against acoustic and vibrational noises. In contrast, the piezo feedback was necessary for compensating the long-term temperature drift of ECDL and keeping the narrow linewidth.

The current feedback signal was added to a laser diode through a bias tee. In order to keep a high gain at low frequencies, several capacitances (0.1 μF ceramic, 1 μF ceramic, two 15 μF tantalum capacitors in series with opposite polarity) were used in parallel as coupling capacitors in the bias tee. A commercial laser diode driver (Thorlabs, LDC202) was coupled to the DC port of the bias tee (coupling inductor: 100 μH) through a RC low pass filter (R: 22

Ω , C: 2.2 mF electrolytic and 10 μ F tantalum in parallel). With this low pass filter, the current noise of the driver was suppressed to lower than other electronic noises arising from a photo detector and a servo circuit. The servo circuit for the current feedback consisted of two stages: the first stage was a normal inverting amplifier and the second stage was a PID amplifier. For enhancing a gain at low frequencies, a slow integrator was added to the positive input of the PID amplifier. This low frequency locking significantly reduced a pedestal below 200 kHz and made locking robust against acoustic and vibrational noises. The feedback bandwidth was limited to 2.4 MHz presumably due to the length of optical fibers (10 m) and cables for the photo-detector signal (6 m) as well as the laser diode itself.

The piezo feedback signal was amplified with a commercial high-voltage amplifier (Matsusada precision, HPZT-0.15PB). Combined with our piezo module (Noliac, CMAP09; capacitance, 440 nF), the amplifier limited the feedback bandwidth to 1 kHz. Since the piezo feedback directly affected the linewidth, the servo circuit for the piezo feedback was obtained by directly integrating the error signal without passing through any amplifier before the integrator. For achieving a linewidth below 100 Hz, it was imperative not to subtract any constant voltage from the error signal for cancelling an offset because then the noise in the constant voltage would be amplified. The offset voltage of the error signal in the PDH method varies with temperature: our solution to cancel the offset will be described hereafter.

As already reported in Ref [11], the offset in the PDH method is due to residual amplitude modulation (RAM) induced by interference effects in the electro-optic modulator (EOM) crystal. It was also pointed out that RAM was significantly attenuated by titling the beam with respect to the crystal by a few degrees. However, the offset still remained in most cases. Since this offset was sensitive to a change in the crystal length, a temperature variation caused a variation in the offset. We found that the remaining offset arose from a mismatch of the light polarization with respect to the crystal. By carefully adjusting the polarization of light before and after the EOM with half waveplates (and sometimes also with quarter waveplates), we could cancel the residual offset to below a thousandth part of the error signal over a few GHz. In order to minimize the fluctuation of polarization, a Glan-Thomson polarizer with an extinction ratio of 1 million was placed before the EOM. In this way, a maximum frequency fluctuation due to RAM was suppressed to below 200 Hz. Further, we surrounded the EOM by a polystyrene foam for passively stabilizing the temperature of the EOM. We confirmed that a frequency fluctuation due to RAM was suppressed to below 10 Hz with our passive method. An active temperature stabilization of the EOM [3] will provide a further stability. Our method had another important advantage that offset-subtraction in a servo circuit was no more required. This allowed us to significantly narrow the linewidth.

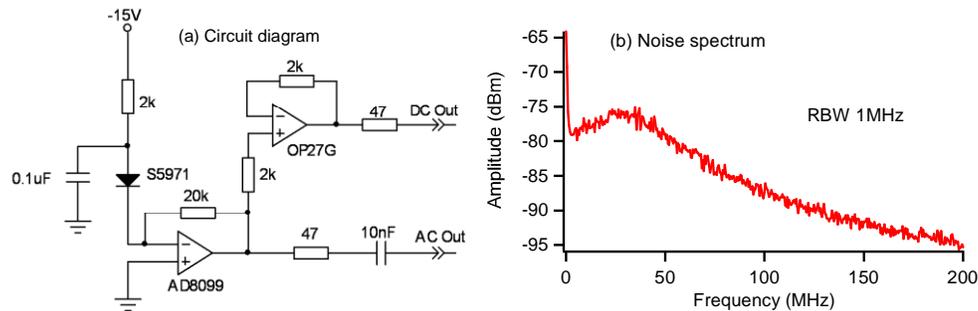


Fig. 5. (a) Circuit diagram of the photo-detector for PDH locking. (b) Noise spectrum of the AC output of the photo-detector. The feedback capacitance required for a proper operation as a transimpedance amplifier is supplied from a parasitic capacitance of the feedback resistance. Instead of adding a feedback capacitance, we added a capacitor and a resistance (2 pF and 51 Ω in series) to the external compensation pin of AD8099 for reducing high frequency noise. By adjusting the compensation capacitance, we could obtain a bandwidth of up to 70 MHz with the same circuit.

The photo-detector for the PDH locking was a home-build transimpedance amplifier composed of Si PIN photodiode (Hamamatsu, S5971), a high-speed op-amp (Analog Devices, AD8099) and a feedback resistance of 20 k Ω (Fig. 5(a)). The DC output was equipped for confirming that locking was successful. Since the noise of the photo-detector should influence the linewidth, we evaluated the noise with a spectrum analyzer (Fig. 5(b)). The increase in the noise below 40 MHz was a result of noise gain, which is the typical behavior of a transimpedance amplifier and is determined by the ratio of the input capacitance (the sum of photodiode and op-amp) to the feedback capacitance. In our case, the noise was dominated by the current noise of AD8099. This photo-detector offered a -3 dB bandwidth of 40 MHz, which was sufficiently higher than our modulation frequency of 17 MHz. The equivalent input noise current at 17 MHz was $0.14 \text{ pA}/\sqrt{\text{Hz}}$. Assuming a photo current of 100 μA , we can obtain a dynamic range of 9 orders, which is sufficient for achieving a linewidth of 1 Hz from the cavity with a linewidth of 200 kHz.

There have been several important points to obtain a narrow linewidth lower than 10 Hz in our setup. Although a part of them have already been reported in Ref [3], we describe all of them below since the situation (especially the finesse of the cavity) was different from Ref [3]. In general, it was important to take care not to cause any interference effects between an optical component and the cavity.

- (1) The phase-modulated light should not pass through an optical fiber. Since the fiber length was sensitive to vibration and temperature variation, an etalon effect in the fiber caused a large variation in the offset of the error signal. In order to avoid this problem, the EOM was placed after the fiber. Further, an optical isolator was placed after the EOM such that reflected light from the cavity did not return to the fiber. We found that at least an isolation of 40dB was required. The reflected light from the cavity was extracted from the input polarizer of the isolator and was used for the PDH locking.
- (2) A photo-detector composed of surface mount components was required for avoiding RF contamination into the photo-detector. A high power RF (17 MHz, 24 dBm) for modulating the light leaked to the ground and contaminated the error signal. Since the amount of the contamination was dependent on the position and shape of coaxial cables for carrying the RF signal, this effect enhanced sensitivity to the vibration. The surface-mounted circuit was also necessary for the proper operation of AD8099.
- (3) Reflection from all of the optics should not hit the photo-detector. When this occurred, interferences between two (or more) reflected lights caused a fluctuation in the offset of the error signal. Especially we had to take care of polarizers and the crystal of the isolator. We used a slightly tilted Glan-Taylor polarizer for combining two lights because it can have a high extinction ratio even if they were slightly tilted. The isolator was also tilted, but then it was difficult to achieve a high transmission. So we removed the input polarizer of the isolator and placed another tilted Glan-Taylor polarizer. We also had to take care so that the transmitted light from the cavity was not scattered or reflected by any other objects.
- (4) Intensity of the light to the cavity should be stabilized to below 0.2% (during a few seconds). We found that an intensity fluctuation due to a polarization fluctuation in the optical fiber caused a frequency fluctuation on the order of 10 Hz. This could be attributed to the asymmetry of the error signal.
- (5) The ground noise should be suppressed as low as possible. The ground of lasers (optical table) should be connected to that of servo circuits with plain stitch copper wires with a cross section of more than 100 mm². In addition, power supplies for electronics (including high power components such as RF amplifiers) should be series regulators; switching regulators should not be used since they add both high frequency (more than 10 kHz) and low frequency (50 Hz) noises on the ground.

With these improvements, we could obtain a linewidth of ~ 1 Hz. Figure 6(a) shows the beat note between two ECDLs at 901 nm independently locked to the neighboring longitudinal mode of the same ULE cavity whereas Fig. 6(b) shows the beat note between a dye laser and an ECDL at 655 nm independently locked to the neighboring longitudinal mode of the same ULE cavity. A slow fluctuation of a few Hz during a several seconds was still remaining because the above problems were not completely circumvented. The regulation of the dye laser was not sufficient because an AOM had a feedback bandwidth of 300 kHz which was determined by the beam diameter in the AOM; hence the linewidth given in Fig. 6(b) should be dominated by the linewidth of the dye laser. Although these measurements could not detect frequency fluctuation due to the vibration of the cavity, we concluded our master lasers had a linewidth narrower than 10 Hz from our acceleration measurement mentioned earlier.

The present approach enables us to obtain narrow linewidths comparable to approaches utilizing a frequency comb, given the frequency comb is locked to a stable reference such as an atomic clock [4, 12]. Without referencing an atomic clock, both approaches are expected to have comparable long-term stabilities, whereas our approach provides narrower linewidths in a short term (~ 1 s) with a simpler setup.

In this study, maximum number of master lasers stabilized to the one end of the cavity was limited to two because two orthogonal polarizations are used for overlapping two lasers. However, in principle, more than two lasers can be independently locked to the cavity if appropriate dichroic mirrors are used for overlapping lasers and color filters are placed before photo-detectors. Thus, limitations in number of lasers originate only from the availability of dichroic mirrors and the cavity mirrors with high finesses at desired wavelengths. A practical limitation in number of available lasers exists in the space on the minus-K optical table: our 600 mm x 600 mm table was completely filled with the optical setup for two colors.

Another important issue is to carry the narrow linewidth of master lasers to slave lasers. The OPLL with a double balanced mixer is an ideal method for realizing it. The details have already given in Ref [13]; hence here we discuss our setup. The beat note between a master and a slave laser was detected via a home-made photo-detector based on a GaAs PIN photodiode with preamplifier (Hamamatsu, G10447-54; bandwidth, 8 GHz; transimpedance, 6 k Ω). The signal from the photo-detector was further amplified by a limiting amplifier (GigOptix, iT3011E; bandwidth, 10GHz; gain, 42 dB). We used a limiting amplifier in order to flatten the frequency response of the photo-detector and enable scanning over a wide frequency range. The signal was integrated after being mixed with a microwave source; for the 901 nm light the signal was feed-backed to the current and the piezo; for the 655 nm light the signal was feed-backed to the AOM and the servo unit of the dye laser. For a diode laser, the bandwidth of the current feedback was 4 MHz, whereas that of the piezo feedback was the same as master lasers (1 kHz). The pedestal was suppressed to lower than -40 dB of the carrier at a resolution bandwidth of 100 kHz. In contrast, there was a significantly larger pedestal in the dye laser than in the diode laser (-30 dB of the carrier at a resolution bandwidth of 10 kHz); this was due to the limited feedback bandwidth of the AOM (~ 300 kHz). The 901 nm light could be continuously scanned for up to 2 GHz, which was limited by a mode-hop free range of the diode laser. The 655 nm light could be continuously scanned for up to 4 GHz, which was limited because the photo-detector signal became smaller at a higher frequency.

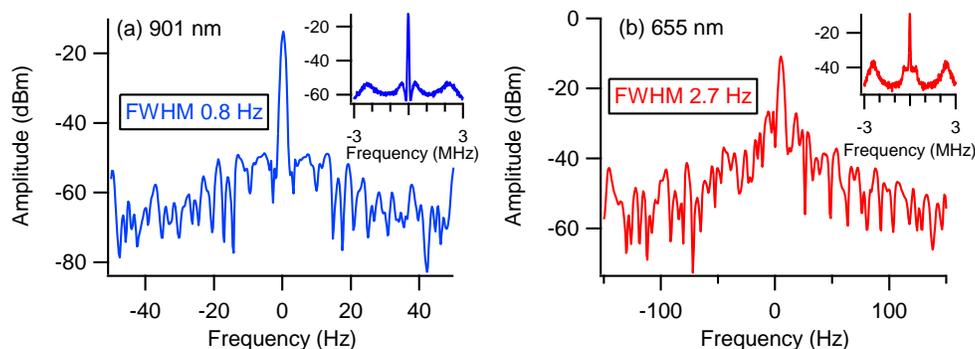


Fig. 6. Beat note spectra between two master lasers locked to a ULE cavity. (a) Two diode lasers at 901nm. (b) A dye and a diode laser at 655 nm. For both wavelengths, spectra in a wide range (6 MHz) are shown in insets.

We also tried to use a digital phase-locked loop circuit (Analog Devices, AD9858) for phase-locking of two diode lasers. In this case, the output from AD9858 was supplied as a DC signal because it was designed to operate a voltage controlled oscillator. However, we finally gave up using it because of three serious problems. First, the bandwidth was limited to ~ 500 kHz and we could not increase it by using a differential gain in the servo circuit because the DC output signal could not be integrated. As a result, the lock was unstable. Second problem was its narrow scanning range (a few 100 MHz) which was determined by the maximum output voltage of 5V supplied from AD9858. The third and the most serious problem was the change in the gain for the current feedback with a change in the DC voltage from AD9858 during a single scan. This was presumably due to the variation in the capacitance of a coupling capacitor before an integrator of a servo circuit, caused by the DC bias characteristics of ceramic capacitors. This problem does not occur for OPLL with a double balanced mixer because a DC voltage required for the piezo feedback is obtained by integrating a small DC signal which does not influence the current feedback.

In summary, we realized a narrow-linewidth two-color light source for 901 nm and 655 nm. Master lasers locked to a ULE cavity, which has finesse of 6000 at 901nm and 10000 at 655nm, provided a linewidth narrower than 10 Hz. Slave lasers locked to master lasers could be continuously scanned over a few GHz with keeping the narrow linewidth. In addition, we could change the wavelength by a few tens of nm since our ULE cavity had coating over a wide range. We have demonstrated that tunability and stability required for STIRAP experiments for molecules was achieved only with a single cavity. In fact, the present setup enabled us to produce ultracold molecules in the rovibrational ground state efficiently [6]. The measurement of the coherence during the two-photon transfer gave an upper limit of the two-photon linewidth of 100 Hz. The present study offers a simple and robust system which is generally applicable to a wide variety of experiments which require a two-color stable light source.

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