All-solid-state tunable continuous-wave ultraviolet source with high spectral purity and frequency stability

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We present a novel approach for the generation of highly frequency-stable, widely tunable, single-frequency cw UV light that is suitable for high-resolution spectroscopy. Sum-frequency generation (SFG) of two solid-state sources with a single cavity resonant for both fundamental waves is employed. Using a highly stable, narrow-linewidth frequency-doubled cw Nd:YAG laser as a master laser and slaving to it the SFG cavity and the other fundamental wave from a Ti:sapphire laser, we generate UV radiation of 33-mW output power around 313 nm. Alternatively, we use a diode laser instead of the Ti:sapphire laser and produce an output power of 2.1 mW at 313 nm. With both setups we obtain a continuous tunability of >15 GHz, short-term frequency fluctuations in the submegahertz range, a long-term frequency drift below 100 MHz/h, and stable operation for several hours. The theory of optimized doubly resonant SFG is also given. © 2002 Optical Society of America

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1. Introduction

The high-resolution spectroscopy of atoms, ions, and molecules often requires practical cw single-frequency lasers that emit in the UV. The traditional solution is a single-frequency dye laser that is resonantly frequency-doubled to the UV.[1] Among numerous examples we mention the works of Bollinger *et al.*[2] and Imajo *et al.*,[3] who produced 313-nm radiation for laser cooling of Be^+. Although such systems are effective and tunable, they are also expensive and maintenance intensive. In a few cases, an all-solid-state approach has been employed where a fixed-wavelength solid-state laser emitting in the near IR (e.g., Nd:YAG at 946 or 1064 nm) is frequency quadrupled in two resonant second-harmonic steps.[4] Although this method simplifies the source aspect, it is only applicable to particular cases.

Another approach to tunable UV generation is a sum-frequency generation (SFG) between two laser sources, of which at least one is frequency tunable. To enhance the conversion efficiency, one or both of the pump waves can be resonated in an enhancement cavity.

Doubly resonant SFG (DR-SFG) of all solid-state lasers was described first by Kaneda and Kubota.[5] The second harmonic of a single-frequency Nd:YAG laser and the fundamental wave of another single-frequency Nd:YAG laser were mixed; up to 0.66 W of 355 nm radiation were obtained. Frequency tuning of the UV light was not investigated. In Kaneda and Kubota's theoretical analysis of DR-SFG, a set of simultaneous equations for the circulating powers was given, from which the optimum cavity parameters and output power can be derived numerically.

The DR-SFG of two Nd:YAG lasers at 1064 and 1319 nm was investigated by Vance *et al.*[6] More than 400 m-W of cw power at a wavelength of 589 nm were generated with a monolithic $LiNbO_3$ resonator. Frequency tuning was not reported.

Recently the development of a tunable single-frequency UV source, based on a Ti:sapphire laser resonantly frequency doubled in a cavity and then sum-frequency mixed with a diode laser in another doubly resonant cavity, was reported.[7] Output powers of 50 mW and tuning around 252 nm were achieved, but fine frequency tuning was not reported.

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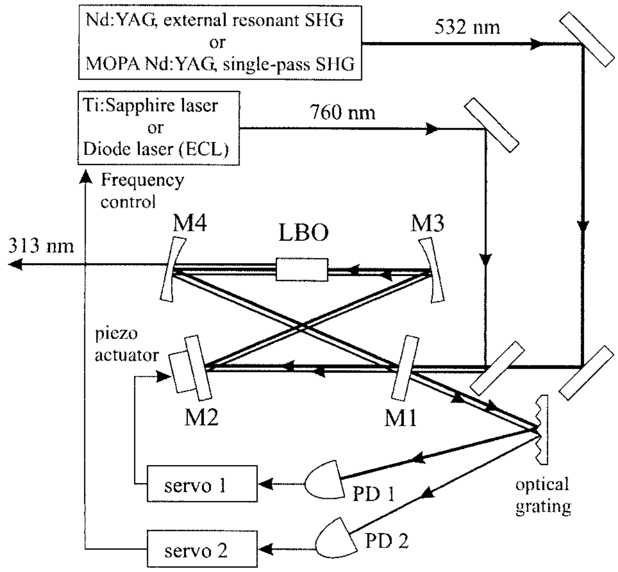

Fig. 1. Schematic of the doubly resonant SFG setup. PD, rf photodetector.

Whereas this approach is flexible and leads to high output powers, it may be too costly for applications where the power requirement is at a level of a few milliwatts.

In this paper we present an all-solid-state scheme that is flexible in terms of UV spectral coverage, has a wide continuous tuning range, and avoids the disadvantages of a dye laser. In particular, we show that if only a few milliwatts UV are needed, the scheme can be implemented with a low-power diode laser, avoiding the costs associated with a Ti:sapphire laser. In addition, we also operate the system with a Ti:sapphire laser to show the generation of higher-output powers. Moreover, we extend the theory of DR-SFG by giving explicit expressions for the optimum incoupling transmissions for reaching the optimum DR-SFG efficiency.

Our approach (shown in Fig. 1) is based on the DR-SFG of single-frequency, doubled Nd:YAG lasers (532 nm) and either a Ti:sapphire or a tunable diode laser. In the second case, where the power delivered by the diode laser is small, the high power available from the Nd:YAG lasers (up to 10 W of single-frequency power at 532 nm are commercially available at present) ensures that the sum-frequency power attains a level that is useful for a wide range of applications. The diode laser can be an external cavity laser or distributed-feedback laser, and is thus of modest cost. Another important feature of our scheme is the excellent absolute frequency stability and the narrow linewidth of the UV light. We achieve these by choosing one of the two fundamental lasers to have high absolute frequency stability and narrow linewidth. When we slave the SFG cavity to this wave and the second laser to the cavity, the spectral properties of the first laser are to a large extent transferred to the UV wave.

2. Theory

We first present general results of the theory of doubly resonant SFG (see also Ref. 5) and derive its optimum performance. Let P_1^{circ}, P_2^{circ} be the powers of the two fundamental waves of angular frequencies ω_1, ω_2 circulating inside the cavity. In the case of the negligible depletion of either wave, the power of the wave $\omega_3 = \omega_1 + \omega_2$ is $P_3 = E_\Sigma P_1^{\text{circ}} P_2^{\text{circ}}$, where E_Σ is the phase-matched SFG nonlinearity,

$$E_\Sigma := 4\,\frac{\mu_0 d^2}{\pi c_0^2}\,\frac{\omega_1\omega_2\omega_3}{n_{\omega_3}^2}\,hL, \tag{1}$$

where d is the effective susceptibility coefficient, L is the crystal length, and h is the Boyd–Kleinman factor. The circulating power P_1^{circ} is given by

$$\left(\frac{T_1 P_1^{\text{in}}}{P_1^{\text{circ}}}\right)^{1/2} = 1 - \left[R_1\left(1 - E_\Sigma\,\frac{\omega_1}{\omega_3}\,P_2^{\text{circ}}\right)\right]^{1/2}, \tag{2}$$

and a corresponding expression holds for P_2^{circ} after subscript exchange $1 \leftrightarrow 2$. P_i^{in} are the incident powers of the two fundamental waves in the TEM_{00} modes matched to the respective cavity modes. $R_i = (1 - T_i)(1 - S_i)$ are the round-trip power reflection coefficients, with T_i being the transmissions of the incoupling mirror, and S_i being the other passive losses at the respective frequencies ω_i.

In general, the intracavity powers are easily found through numerical solution of the two coupled Eqs. (2) (obtained by subscript exchange $1 \to 2$), and P_3 then is calculated. The operating point for maximum SFG power is of particular interest; it occurs when the incoupling transmissions T_i are chosen (for given values of input powers, losses, and nonlinearity) so that both waves are impedance matched into the cavity. The optimum values can be calculated explicitly in the case of small $S_i \ll 1$ and small internal efficiencies $E_\Sigma P_i^{\text{circ}} \ll 1$ when the square root in the right side of Eq. (2) can be approximated. Defining the normalized powers ϵ_i and the function $f(\epsilon_1, \epsilon_2)$ as

$$\epsilon_1 := \frac{\omega_2}{\omega_3}\,\frac{E_\Sigma P_1^{\text{in}}}{S_1 S_2}, \qquad \epsilon_2 := \frac{\omega_1}{\omega_3}\,\frac{E_\Sigma P_2^{\text{in}}}{S_1 S_2}, \tag{3}$$

$$f := 1 + \epsilon_1 + \epsilon_2 + [(1 + \epsilon_1 + \epsilon_2)^2 - 4\epsilon_1\epsilon_2]^{1/2}, \tag{4}$$

we have the maximum output power

$$P_3^{\max} = \frac{\omega_3^2}{\omega_1\omega_2}\,\frac{S_1 S_2}{E_\Sigma}\,\frac{\epsilon_1\epsilon_2}{f/2}, \tag{5}$$

for the optimum choices $T_i^{\text{opt}}/S_i = f/2 - \epsilon_i$. We see that the maximum output is crucially dependent on the product E_Σ/S_1S_2. In the regime of low-input powers, $\epsilon_i \ll 1$, we have the simple result $P_3^{\max} = E_\Sigma P_1^{\text{in}} P_2^{\text{in}}/(S_1 S_2)$. In the limit of one weak and one strong fundamental wave, for example $\epsilon_1 \gg 1$, $\epsilon_2 \ll 1$, we have full conversion of the weak wave, $P_3^{\max} = \omega_3 P_2^{\text{in}}/\omega_2$.

3. Experiment

Our experimental implementation of DR-SFG is designed for generation of 313-nm radiation for laser cooling of trapped Be^+.

In our first experimental setup, capable of delivering 33-mW UV radiation at 313-nm wavelength, we use a resonantly frequency-doubled monolithic Nd:YAG laser[8,9] together with a Ti:sapphire laser as fundamental lasers. The Nd:YAG laser (InnoLight, Diabolo) produces up to 1 W at 532 nm with a narrow linewidth of <100 kHz, a frequency drift of <100 MHz/h, and a continuous tuning range of approximately 12 GHz. The Ti:sapphire laser (Coherent, MBR-110) is pumped by a frequency-doubled Nd:YAG laser (Coherent, Verdi-V10) and produces up to 1.5 W at 760-nm wavelength with a linewidth of about 100 kHz, a coarse tuning range of 80 nm (with the particular optics set in use), and a mode-hop free-tuning range of approximately 30 GHz.

In our second experimental setup, aimed at demonstrating a low-cost alternative setup based on a Ti:sapphire laser, we use an external-cavity diode laser as one of the two fundamental lasers. We also use a different 532-nm laser, a frequency-doubled master oscillator–power amplifier (MOPA) Nd:YAG laser (Lightwave Electronics 6000). The MOPA Nd:YAG laser is single-pass frequency-doubled in a 60-mm-long periodically poled lithium niobate crystal, fabricated at the University of Kaiserslautern. 8 W of Nd:YAG light at 1064 nm produce up to 3.2 W at 532 nm, which corresponds to 40% single-pass conversion efficiency. The high power of the MOPA Nd:YAG laser ensures that the sum-frequency power attains a level useful for a wide range of applications. The frequency-doubled light at 532-nm wavelength inherits a narrow linewidth of about 10 kHz, a frequency drift <20 MHz/h, and a high mode quality ($TEM_{00}, M^2 < 1.1$) from the 1064-nm Nd:YAG master laser. By changing the temperature of the Nd:YAG crystal we can frequency tune the master laser by 8 GHz without mode hops. The external-cavity diode laser (TUI Optics, DL100) has a maximum output power of approximately 12 mW. The Littrow configuration gives a coarse tuning range of 6 nm, and the optical feedback into the laser diode guarantees a small linewidth of <1 MHz. Control of grating position and diode current allows continuous tuning by up to 15 GHz.

The sum frequency is generated in a bow-tie-shaped ring cavity that is doubly resonant for 532 and 760 nm and contains a $L = 15$-mm-long lithium triborate (LBO) crystal with triple antireflection coatings ($R^c_{532} \approx 0.7\%$, $R^c_{760} \approx 0.2\%$, $R^c_{313} < 2\%$). The resonator consists of an incoupling mirror M1 and three mirrors M2–M4, each with a highly reflective coating for 532 and 760 nm and an antireflective-coating for 313 nm. Mirror M2 is mounted on a piezo actuator; M3 and M4 are concave mirrors with $r = -100$ mm. This gives a focus of the cavity mode at the center of the crystal of $w_{c,532} = 41$ μm and $w_{c,760} = 49$ μm. The astigmatism is about 4%. The cavity length is 483 mm. The LBO crystal is cut for type-I ooe interaction in the xy plane with $\varphi = 54.25°$, yielding $n_{\omega_3} = 1.618$, and $d = d_{32} \cos \varphi \approx 0.5$ pm/V. The walk-off angle due to birefringence is $\rho = 1.2°$. The Boyd–Kleinman factor, including the walk-off effect, is estimated to be $h \approx 0.1$ (Ref. 10). With these values, the SFG nonlinearity can be calculated according to Eq. (1) as $E_\Sigma = 0.138$/kW.

The transmissions of the incoupling mirror are $T_{532} = 1.6\%$ and $T_{760} = 2.7\%$. By locking the SFG cavity in turn to each fundamental wave, while the other fundamental wave was off, we determined enhancement factors of 15 and 54 at 532 and 760 nm, respectively. According to Eq. (2) this gives passive round-trip losses of $S_{532} = 4.8\%$ and $S_{760} = 1.8\%$. These values show that the cavity is undercoupled for the 532-nm wave, and overcoupled for the 760-nm wave (even when taking the conversion losses into account). A comparison between experimental and theoretical conversion efficiency was performed for both experimental setups. In the first setup, based on the Ti:sapphire laser, the available fundamental powers are $P_1^{in} = 425$ mW at 532 nm and $P_2^{in} = 492$ mW at 760 nm. These are the powers in the modes matched to the resonator modes. The mode-matching factors were 95.5% and 84.1% at 532 and 760 nm, respectively, determined through measurement of the strength of higher-order modes when scanning the SFG cavity. Numerical solution of Eq. (2) yields an expected UV power of $P_3 = 22$ mW. Experimentally we obtained 33 mW, which is 50% more than the theoretical value. In the second experimental setup, based on the diode laser, the available mode-matched fundamental powers were $P_1^{in} = 1.23$ W at 532 nm and $P_2^{in} = 5.44$ mW at 760 nm. The mode-matching factors were 87.6% and 47.3% at 532 and 760 nm, respectively. Experimentally, we obtained an UV power of 2.1 mW, which is approximately 90% more than the theoretically expected UV power of $P_3 = 1.1$ mW.

According to Eqs. (3)–(5) the optimum transmissions of the input coupler in our first (Ti:sapphire) setup would be $T_{532}^{opt} = 5.1\%$ and $T_{760}^{opt} = 1.8\%$ (both close to the respective losses S_{532} and S_{760}). With these optimum transmissions the theoretically expected UV power increases significantly (by 40%) to $P_3^{max} = 31$ mW. Optimization of the incoupling transmissions in our second (diode laser) setup, including the diode laser, would predict only a minor increase of the theoretically expected UV power by 10% to $P_3^{max} = 1.2$ mW.

It should be mentioned that the above values for the measured UV powers are stable values, reached after some minutes of operation of the SFG system. For the first (Ti:sapphire) setup, for approximately 10 s after locking the SFG cavity to the 532-nm wave and the 760-nm wave to the cavity, the system produced approximately 48-mW UV output power, which then dropped to 33 mW within a few minutes. Thermal effects in the LBO crystal could be the reason for the observed power changes.

A feature of our system is that in order to obtain

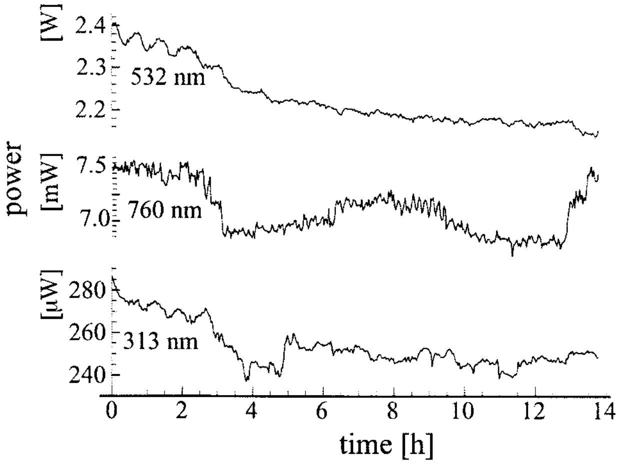

Fig. 2. Input fundamental and output SFG powers during stable long-term operation.

spectrally pure and frequency-stable 313-nm light that is suitable for high-resolution spectroscopy, it is sufficient if one of the two fundamental lasers has good spectral properties. When we slave the doubly resonant cavity to this laser and slave the second fundamental laser to the cavity, the spectral properties of the first fundamental laser can be transferred to a large extent to the generated UV light.

The frequency locks used for stabilizing the system all employ the phase-modulation technique[11] in reflection (Fig. 1). In our first (Ti:sapphire) setup, where both fundamental lasers have similar good spectral properties, either of the two lasers would be suitable as a master laser for the SFG system. We have chosen to lock the cavity to the 532-nm light. To this end, we used the 12-MHz phase modulation of the 1064-nm Nd:YAG laser by an electro-optical modulator, which is required to stabilize the external frequency-doubling cavity to the 1064-nm wave. A 400-kHz phase modulation was imparted to the Ti:sapphire laser by way of a piezo-mounted cavity mirror. In our second (diode laser) setup, the ultranarrow linewidth and high intrinsic frequency stability of the 1064-nm master laser in the MOPA system predestines the MOPA to be the master laser of the SFG system. To this end, the 1064-nm laser in the MOPA system is phase modulated at 3.2 MHz by way of a piezotransducer on the Nd:YAG crystal. Phase modulation of the diode laser frequency is obtained through modulation of the diode current at 20 MHz by way of a bias tee. The waves reflected from the incoupling mirror are separated by a grating and are detected by rf photodetectors. The cavity length is stabilized by using the 532-nm error signal. The servo (servo 1) has 10-kHz bandwidth. The laser-diode servo (servo 2) that stabilizes the laser diode frequency to the cavity has a slow channel with 3-kHz bandwidth acting on the external grating and a fast channel of 200-kHz bandwidth acting on the diode current. The locks are stable and allow uninterrupted operation for many hours, as shown in Fig. 2.

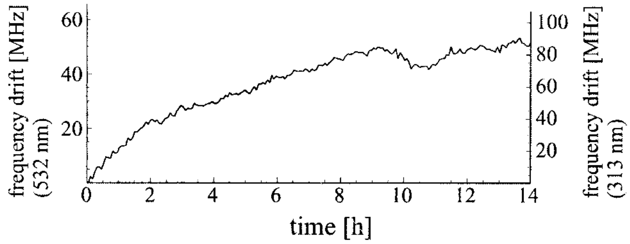

Fig. 3. Frequency drifts of the 532- and 313-nm light.

The 15% drift of the UV power is mainly due to power drifts of the fundamental waves caused by environmental changes. The low-frequency drift of the MOPA Nd:YAG laser leads to a correspondingly low drift of the UV frequency. Figure 3 shows the measured frequency drift of the MOPA Nd:YAG laser from which a UV drift rate of less than 20 MHz/h can be inferred. If a still-lower drift is required, the frequency-doubled Nd:YAG light could be locked to a hyperfine transition in molecular iodine. A drift rate of less than 200 kHz/h should be achievable.

With the locking scheme employed, a frequency change $\Delta\omega_1$ of the "master" wave causes a frequency change $\Delta\omega_2/\omega_2 = \Delta\omega_1/\omega_1$ of the other fundamental wave. This holds if drifts in the two servos and the influence of the crystal are neglected. Figure 4 shows the response of the locked-diode laser at 760 nm to a tuning of the 532 nm wave of the frequency-doubled MOPA Nd:YAG laser. The tuning is found to be $\Delta\nu_{760} = 0.6997(4)\Delta\nu_{532}$, in agreement with the expected value of 0.69973. The servo errors are estimated to lie well below 1 MHz on time scales of 1 h. Thus the UV sum frequency essentially is controlled by the frequency of the MOPA Nd:YAG laser, and the total tuning is given by $\Delta\nu_{313} = \Delta\nu_{532} + \Delta\nu_{760} = 1.6997\Delta\nu_{532}$. Figure 4 shows that the diode laser SFG system allows 16 GHz of continuous tuning in the UV without mode hops. With the Ti:sapphire SFG system we achieved a comparable continuous tuning range of slightly more than 15 GHz, whereby a sweep through the whole tuning range was possible in about 1 s.

As a further characteristic of the laser system, we consider the frequency fluctuations of the generated

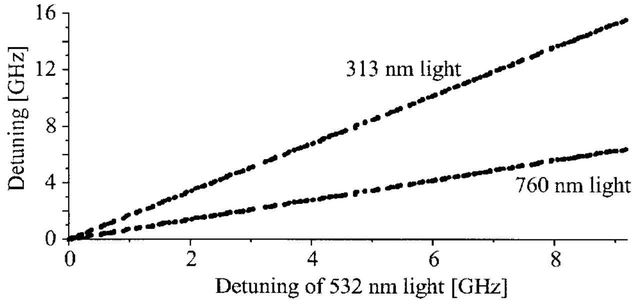

Fig. 4. Frequency tuning of the UV light. Frequencies of the 532-nm light and the slaved 760-nm light were measured simultaneously with wavemeters.

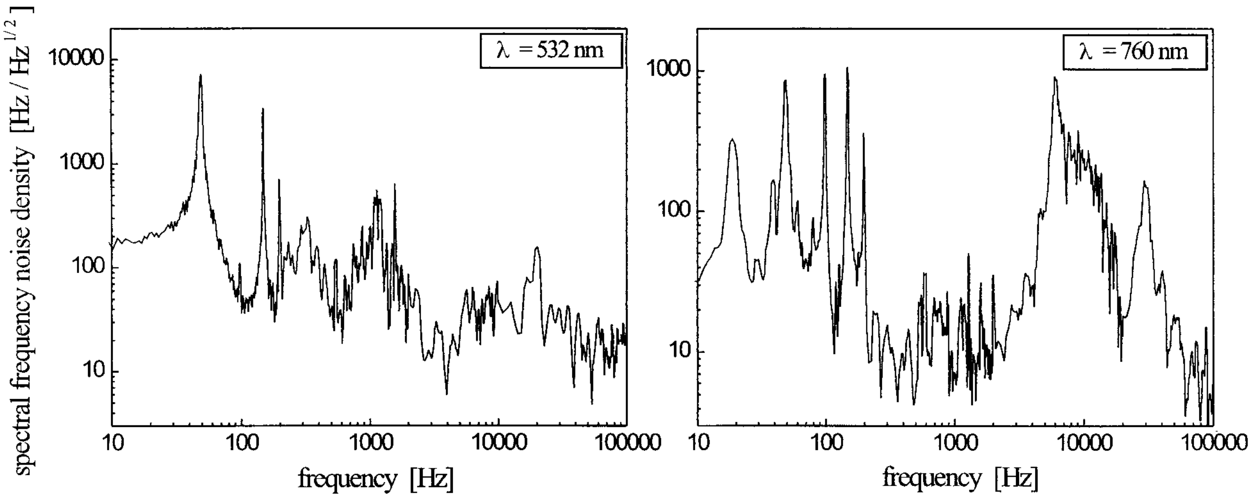

Fig. 5. Spectral frequency noise densities of the cavity lock error signals. 532-nm Nd:YAG laser (left) and 760-nm Ti:sapphire laser (right).

UV light, which are indicative of the linewidth. The UV frequency fluctuations $\delta\nu_{313}(t)$ are caused by the frequency fluctuations $\delta\nu_{532}^{ML}(t)$ of the 532-nm master laser and the frequency fluctuations $\delta\nu_{760}^{SL}(t)$ of the 760-nm second fundamental slave laser. Since the frequency of the latter is controlled by the frequency of the 532-nm master laser by way of the two frequency locks, the frequency fluctuations $\delta\nu_{760}^{SL}(t)$ can be expressed in terms of the deviations $\delta\nu_{532}^{cav}(t)$ and $\delta\nu_{760}^{cav}(t)$ between the SFG cavity resonance frequencies at 532 and 760 nm and the respective laser frequencies. The reason for these deviations are nonideal frequency locks due to finite bandwidths, offset errors, electronic noise, etc. We can write

$$\delta\nu_{313}(t) = \left(1 + \frac{532}{760}\right)\delta\nu_{532}^{ML}(t) - \left(\frac{532}{760}\right) \times \delta\nu_{532}^{cav}(t) + \delta\nu_{760}^{cav}(t). \tag{6}$$

Equation (6) shows that with a sufficient tight lock [i.e., small $\delta\nu_{532}^{cav}(t)$, $\delta\nu_{760}^{cav}(t)$], the UV frequency fluctuations can in principle be reduced to the level of those of the master laser. Note that since a fast reaction of the cavity-to-master-laser frequency lock to an external perturbation will also result in a readjustment of the second fundamental-laser-to-cavity frequency lock, there will always be a certain degree of correlation between $\delta\nu_{532}^{cav}(t)$ and $\delta\nu_{760}^{cav}(t)$.

To determine the rms frequency fluctuations of the generated UV light, we took time records of the error signals of both frequency locks over 1 s within a bandwidth of 100 kHz. The error signals are direct measures of the deviations $\delta\nu_{532}^{cav}(t)$ and $\delta\nu_{760}^{cav}(t)$. For the Ti:sapphire SFG setup, we have measured rms values of $[\langle(\delta\nu_{532}^{cav})^2\rangle]^{1/2} = 24$ kHz and $[\langle(\delta\nu_{760}^{cav})^2\rangle]^{1/2} = 55$ kHz. The respective spectral frequency noise densities are shown in Fig. 5. The frequency fluctuations of the (free-running) Nd:YAG master laser in the same bandwidth as above are approximately $[\langle(\delta\nu_{532}^{ML})^2\rangle]^{1/2} = 50$ kHz. Assuming the (unrealistic) worst case of complete correlation between $\delta\nu_{532}^{ML}(t)$, $\delta\nu_{532}^{cav}(t)$, and $\delta\nu_{760}^{cav}(t)$, one can calculate an upper limit for the rms value of the UV frequency fluctuations of $[\langle(\delta\nu_{313})^2\rangle]^{1/2} < 160$ kHz.

For the diode-laser SFG setup, the frequency fluctuations of the diode laser are several orders of magnitude greater than the frequency fluctuations of the MOPA Nd:YAG master laser, which are known from previous measurements to be approximately 5 kHz (Ref. 12). For this setup, a tight lock is the essential tool used to reduce the linewidth of the generated UV light. With the servo system used for the diode laser, which features a fast channel of 200-kHz bandwidth acting on the diode current, we estimate that the UV frequency fluctuations within a bandwidth of 100 kHz have a rms value well below 1 MHz.

4. Conclusion

We have presented the theory and experimental implementation of doubly resonant sum-frequency generation in a single cavity using two different combinations of lasers as fundamental sources. The first combination, suitable to produce high-UV output powers, is based on a resonantly frequency-doubled monolithic Nd:YAG laser and a Ti:sapphire laser. Frequency-tunable cw UV light with a high long-term frequency stability (drift <100 MHz/h) and short-term frequency fluctuations at the 100 kHz level has been produced with 33-mW power. The second combination, which avoids the costs associated with a Ti:sapphire laser, is based on a diode laser and a frequency-doubled MOPA Nd:YAG laser. Frequency-tunable UV light with a high long-term frequency stability (drift <20 MHz/h) and short-term frequency fluctuations in the submillihertz range has been produced with 2.1-mW power. The large continuous tuning range of >15 GHz, achievable with both combinations of fundamental lasers, is of particular importance, e.g. for laser cooling in rf-traps and for resolving hyperfine structures. The features of the described approach and the large UV wavelength coverage achievable, together with the possibility of using readily available diode lasers, should make it of significant interest to spectroscopists.

We have implemented an absolute frequency stabilization for the first setup by locking the 532-nm Nd:YAG laser to a hyperfine transition of molecular iodine. To this end, a fraction of the 532-nm wave was split off, frequency shifted by an acousto-optic modulator in a double-pass configuration, and sent into a compact setup for Doppler-free modulation transfer spectroscopy. The error signal was fed back to the Nd:YAG laser for stabilization. The optical frequency of the main laser wave could be tuned by up to 200 MHz by changing the acousto-optic modulator frequency shift. We thus obtained absolutely stabilized UV radiation (frequency instability < 1 MHz/h), tunable over a range of 340 MHz.

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