Tunable source of terahertz radiation using molecular modulation

D. D. Yavuz* and J. J. Weber

Department of Physics, 1150 University Avenue, University of Wisconsin at Madison, Madison, Wisconsin 53706, USA
**Corresponding author: yavuz@wisc.edu*

Received August 7, 2012; revised September 3, 2012; accepted September 5, 2012;
posted September 5, 2012 (Doc. ID 174005); published October 3, 2012

We describe a source of terahertz (THz) radiation that is based on Raman down-shifting of an infrared laser beam using highly coherent molecular vibrations. The source can operate in either the pulsed or the continuous wave (CW) regime and is tunable over much of the THz region of the spectrum (1–10 THz). In the pulsed regime, we predict average output powers of order 10 mW and peak powers approaching 1 MW. In the CW regime, average powers exceeding 100 μW with spectral linewidths at the hertz level are achievable. © 2012 Optical Society of America

OCIS codes: 190.5650, 290.5910, 140.3070.

The terahertz (THz) region of the electromagnetic spectrum is sometimes referred to as the *terahertz gap*, as it has proven difficult to develop light sources that produce these frequencies. The interest in THz radiation sources has been continually growing over the last two decades due to an extensive list of potential applications in a variety of fields including remote sensing and imaging [1,2]. Although significant advances have been made in recent years, obtaining a high power and widely tunable coherent source of THz radiation remains a very challenging task. In this Letter, we propose to use the technique of *molecular modulation*, which utilizes four-wave mixing in a coherently prepared molecular gas, for THz wave generation [3]. As shown in Fig. 1(a), two intense laser beams, called the pump (E_P) and the Stokes (E_S), drive a selected molecular Raman transition. If the two driving laser beams are sufficiently intense, one can coherently transfer almost half of the population from the ground level $|1\rangle$ to the excited level $|2\rangle$, and approach a state of maximum coherence (off-diagonal density matrix element $\rho_{12} = 1/2$). With the molecules coherently prepared, a third longer-wavelength laser, called the mixing beam (E_M), mixes with the coherent vibrations and produces the frequency-downshifted THz wave (E_T). As we discuss below, the peak output power and the tuning range of our proposed THz source far exceeds the capabilities of existing devices [1,2].

Molecular modulation has recently evolved into a powerful technique for generating broadband coherent optical spectra [4]. Experiments have traditionally been performed using Q-switched pulsed pump and Stokes lasers that are loosely focused on a low-pressure molecular gas cell. Preparation of near-maximal molecular coherence requires incident laser intensities of order 1 GW/cm^2. With the molecules prepared, the driving lasers can mix with the established molecular coherence and produce many Stokes and antiStokes orders. Harris and Hakuta groups have produced broadband Raman spectra covering much of the optical region using this approach [3,5]. A portion of the produced broad spectrum may be phase-locked using spectral modification techniques, producing optical pulses that are less than a single cycle in duration [6]. Recently, Kung and colleagues have demonstrated the first nonsinusoidal waveform synthesis using the molecular modulation technique [7]. The extension of this technique to the CW domain requires either hollow-core photonic crystal fibers with small mode area or high-finesse cavities [8,9]. In the cavity approach, CW pump and Stokes laser beams are locked to the longitudinal modes of the gas-filled cavity to produce high intracavity circulating intensities that establish a CW molecular coherence [10]. Recently, the synthesis of near-single-cycle pulses using CW spectral components has been demonstrated using this approach [11].

When the driving laser beams are opposite circularly polarized, the generation of higher-order Stokes and anti-Stokes beams is suppressed due to angular momentum selection rules. In this case, an appropriately polarized mixing beam can be frequency up-shifted or down-shifted, and thus the molecular medium may be utilized as an efficient frequency converter [12]. The mixing process is linear in the mixing beam intensity and as a result the mixing beam can be arbitrarily weak. As shown in Fig. 1(a), if the

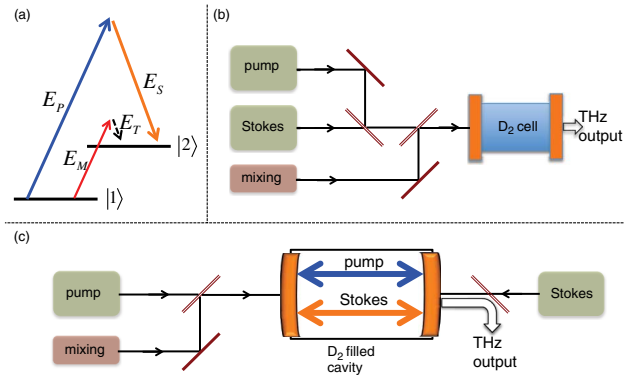

Fig. 1. (Color online) (a) Energy level diagram of our proposal. Intense pump (E_P) and Stokes (E_S) beams prepare the molecules into a highly coherent state. The established coherence then mixes with a separate infrared mixing beam (E_M) to produce the long-wavelength THz radiation (E_T). (b) Pulsed THz source. Q-switched pump, Stokes, and mixing laser beams are loosely focused to a Raman cell in order to produce the THz wave. (c) CW THz source. The molecules are placed in a cavity with a high finesse at the pump and the Stokes wavelengths. The mixing beam passes through the system only once and produces the CW THz wave.

frequency of the mixing beam is close to the Raman transition frequency, the generated beam is long-wavelength THz radiation. In this Letter, our goal is to analyze this approach in detail for realistic experimental parameters for two different schemes. In the scheme of Fig. 1(b), Q-switched pulsed pump and Stokes laser beams drive the molecular coherence. A separate pulsed mixing beam is then frequency down-shifted to produce a pulsed THz source. We also consider the CW scheme of Fig. 1(c) in which CW pump and Stokes beams establish a CW molecular coherence inside a high-finesse cavity. A CW mixing beam that is not resonant with the cavity (i.e., the mirrors do not have a high reflectivity at the mixing laser wavelength) is down-shifted in a single pass to produce the CW THz wave.

We proceed with a detailed description of our suggestion. We follow closely the formalism of Harris and Sokolov [13]. Due to the long wavelength of the THz radiation, the diffractive effects are important, and we consider the generation of the THz wave in the full three spatial dimensions. We assume near-monochromatic excitation and work with the slowly varying envelope $E_T(x,y,z)$ such that the total field is $\hat{E}_T(x,y,z,t) = \mathrm{Re}\{E_T(x,y,z)\exp[j(\omega t - kz)]\}$, where ω is the angular frequency and $k = \omega/c$. The molecular coherence is established with the intense pump and Stokes laser beams, and we take the mixing beam to be sufficiently weak so that it does not interfere with the coherence preparation. Due to low conversion efficiency, we ignore the depletion of the mixing laser as a result of the THz wave generation. For the THz beam driven by the dipole moment induced in the medium, the slowly varying-envelope propagation equation is [14]

$$\frac{\partial E_T}{\partial z} + \frac{j}{2k}\left(\frac{\partial^2 E_T}{\partial x^2} + \frac{\partial^2 E_T}{\partial y^2}\right) = -j\eta\hbar\omega N(a\rho_{11}E_T + d\rho_{22}E_T + b\rho_{12}E_M), \quad (1)$$

where $\eta = \sqrt{\mu_0/\epsilon_0}$ and N is the molecular density. The quantities ρ_{11} and ρ_{22} are the populations of the two Raman levels (the diagonal density matrix elements). The constants a, d, and b determine the refractive index of the gas and the Raman polarizability. These constants are calculated by summing through the many ro-vibrational levels in the excited electronic configuration with corresponding matrix elements and detunings [13]. We take the molecular system to be prepared adiabatically to a state that is smoothly connected to the ground state. This preparation can be accomplished by utilizing a small two-photon detuning from the Raman resonance, $\delta\omega \equiv (\omega_1 - \omega_2) - (\omega_P - \omega_S)$. The driving two-photon Rabi frequency for the Raman excitation is $B = b_P E_P E_S^*$, where b_P is the Raman polarizability at the pump wavelength. We define $B = |B|\exp(j\varphi)$ and $\tan\theta = |B|/(\delta\omega + A/2 - D/2)$. The quantities $A/2$ and $D/2$ are the Stark shifts of the Raman levels $|1\rangle$ and $|2\rangle$ due to the intense pump and Stokes beams. With these definitions, the adiabatic solutions for the density matrix elements of the Raman transition are

$$\rho_{11} = \cos^2\left(\frac{\theta}{2}\right); \qquad \rho_{22} = \sin^2\left(\frac{\theta}{2}\right), \qquad \rho_{12} = \left(\frac{1}{2}\sin\theta\right)\exp(j\varphi). \quad (2)$$

In our numerical simulations, we calculate the conversion efficiency from the mixing beam to the generated THz wave by numerically integrating Eq. (1) together with the adiabatic solution of Eq. (2) on a three-dimensional $x-y-z$ grid. We consider the $|\nu = 0, J = 0\rangle \rightarrow |\nu = 1, J = 2\rangle$ ro-vibrational transition in molecular deuterium (D2) at a transition frequency of 3171 cm^{-1} (95 THz). We take the molecules to be cooled to the rotational ground state by, for example, immersing the cell in a liquid-nitrogen bath. We take the pump and the Stokes laser beams to be at wavelengths of 1.04 and 1.55 μm. The mixing beam wavelength is tuned from 3.1 to 2.86 μm to tune the generated THz radiation over the frequency range 1–10 THz. We analyze THz generation with realistic experimental conditions for two scenarios: pulsed [Fig. 1(b)] and CW [Fig. 1(c)]. For both cases, we take the pump, the Stokes, and the mixing beams to have Gaussian spatial profiles with beam waists of 1 mm. We take into account the refractive index and dispersion of the molecular gas for all the beams and therefore include the effects of phase mismatch as the THz wave is generated along the cell. We take the cell length to be $L = 50$ cm and assume $N = 2.69\times10^{19}/\mathrm{cm}^3$ (1 amagat number density). We note that the conversion efficiency is maximized for a maximally coherent state. Well before this limit is achieved, the efficiency roughly scales as $I_P^2 I_S^2 N^2 L^2$ where I_P and I_S are the intensities of the two driving lasers.

Figure 2(a) shows the conversion efficiency from the mixing beam to the THz beam for the pulsed source. We take both the pump and the Stokes laser beams to have a pulse energy of 1 J with a typical Q-switched pulse duration of 10 ns. The peak power is 100 MW, and the peak intensity at the focus is 6.36 $\mathrm{GW/cm}^2$ for both laser beams. We take the two-photon detuning to be $\delta\omega = 2\pi\times0.5$ GHz. For these parameters, the molecular coherence achieved at the focus is $\rho_{12} = 0.3$. The conversion efficiency from the mixing beam to the generated beam varies from 3×10^{-5} to 2×10^{-2} as the generated frequency is tuned from 1 to 10 THz. The efficiency increases with increasing THz frequency for two reasons: (i) the frequency factors in the propagation equation of Eq. (1), and (ii) there is slower diffraction of the THz

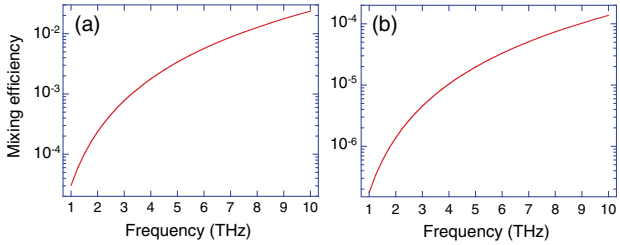

Fig. 2. (Color online) Conversion efficiency from the mixing beam to the generated THz beam for (a) pulsed and (b) CW molecular excitation. The simulation is performed by assuming experimental parameters that are currently achievable using state-of-the-art laser technology. See text for details.

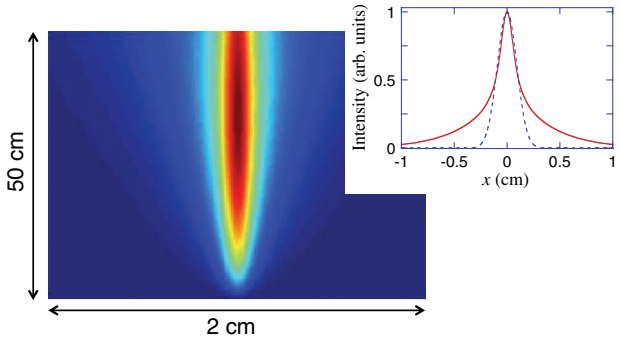

Fig. 3. (Color online) THz intensity along one of the transverse coordinates $I_T(x, y = 0, z)$. The inset shows the intensity at the end of the cell (the top of the two-dimensional false-color plot) as a function of one of the transverse coordinates. For comparison, a Gaussian intensity profile with the same width is also plotted (dashed line).

wave at higher frequencies. If we assume a mixing beam with modest parameters of 100 mJ per pulse at a 10 Hz repetition rate, such a source would produce an average power of 30 μW to 20 mW, with a peak power of 0.3 kW to 0.2 MW, as the output is tuned from 1 to 10 THz.

Figure 2(b) shows the conversion efficiency to the THz beam for a CW experiment [11]. We assume 200 W pump and Stokes laser beams (which are commercially available using ytterbium and erbium-doped fiber lasers) and mirror reflection coefficients of 99.995% for the cavity mirrors. For these parameters, when the lasers are locked to the cavity, the intracavity circulating power for each laser is 4 MW and the circulating intensity at the focus is 255 MW/cm^2. We note that Meng *et al.* have demonstrated CW optical damage threshold exceeding 100 MW/cm^2 for high-quality dielectric coatings [15]. We take the two-photon detuning to be $\delta\omega = 2\pi\times$ 0.25 GHz. The molecular coherence that is achieved at the focus is $\rho_{12} = 0.024$. The conversion efficiency varies from 1.8×10^{-7} to 1.4×10^{-4} as the output is tuned from 1 to 10 THz. A 1 W mixing beam would therefore produce an output power of 0.2 μW to 0.1 mW as the output frequency is tuned. We note that the linewidth of the generated THz wave is determined by the linewidth of the interacting lasers, and not by the linewidth of the Raman transition. Because the pump and Stokes beams are locked to the longitudinal modes of a high-finesse cavity, their linewidths can be made very narrow. This means that the linewidth of the molecular oscillations can also be made very narrow. As a result, if a frequency stabilized mixing laser beam is used, it is, in principle, possible to generate THz radiation with 1 Hz level linewidths.

Figure 3 shows a two dimensional false-color plot of the intensity for a 1 THz wave being generated inside a cell. The inset shows the intensity profile at the output of the cell (solid line). For comparison, a Gaussian intensity profile with the same width is also plotted (dashed line). Due to the high divergence of the THz wave, the output beam is not Gaussian but instead reveals a cusp structure.

We thank Zach Simmons, Dan Sikes, and Jared Miles for many helpful discussions. This work has been supported by the National Science Foundation (NSF).

References

1. M. Tonouchi, Nat. Photonics **1**, 97 (2007).
2. M. Scheller, J. M. Yarborough, J. V. Moloney, M. Fallahi, M. Koch, and S. W. Koch, Opt. Express **18**, 27112 (2010).
3. A. V. Sokolov, M. Y. Shverdin, D. R. Walker, D. D. Yavuz, A. M. Burzo, G. Y. Yin, and S. E. Harris, J. Mod. Opt. **52**, 285 (2005).
4. S. Yoshikawa and T. Imasaka, Opt. Commun. **96**, 94 (1993).
5. J. Q. Liang, M. Katsuragawa, F. Le Kien, and K. Hakuta, Phys. Rev. Lett. **85**, 2474 (2000).
6. W. Chen, Z. Hsieh, S. Huang, H. Su, T. Tang, C. Lin, C. Lee, R. Pan, C. Pan, and A. H. Kung, Phys. Rev. Lett. **100**, 163906 (2008).
7. H. S. Chan, Z. M. Hsieh, W. H. Liang, A. H. Kung, C. K. Lee, C. J. Lai, R. P. Pan, and L. H. Peng, Science **331**, 1165 (2011).
8. F. Couny, F. Benabid, and P. S. Light, Phys. Rev. Lett. **99**, 143903 (2007).
9. J. K. Brasseur, K. S. Repasky, and J. L. Carlsten, Opt. Lett. **23**, 367 (1998).
10. D. D. Yavuz, Phys. Rev. A **76**, 011805(R) (2007).
11. J. T. Green, J. J. Weber, and D. D. Yavuz, Phys. Rev. A **82**, 011805(R) (2010).
12. A. V. Sokolov, Scott Sharpe, M. Y. Shverdin, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Opt. Lett. **26**, 728 (2001).
13. S. E. Harris and A. V. Sokolov, Phys. Rev. A **55**, R4019 (1997).
14. M. Y. Shverdin, D. D. Yavuz, and D. R. Walker, Phys. Rev. A **69**, 031801(R) (2004).
15. L. S. Meng, J. K. Brasseur, and D. K. Neumann, Opt. Express **13**, 10085 (2005).