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Continuous-wave light modulation at molecular frequencies

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By using continuous-wave (CW)–stimulated Raman scattering inside a high-finesse cavity, we generate three CW spectral components covering about one octave of optical bandwidth. We investigate the mutual coherence of these three beams by studying phase-dependent second harmonic generation. From the high contrast of the observed interference fringes, we infer very good phase coherence across the spectrum and thereby infer the synthesis of a near single-cycle optical wave form.

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It is well known that when light is modulated, it typically produces three equally spaced frequencies: one carrier and two sidebands. These frequencies can be used, for example, to synthesize frequency-modulated (FM) or amplitude-modulated (AM) temporal wave forms, depending on their relative phase. Light can be modulated at rates exceeding 10 GHz using nonlinear optical effects such as electro-optic light modulation. Modulating light at much higher rates has proved to be difficult. It was only in 2001, more than four decades after the invention of the laser, that light modulation at molecular frequencies was demonstrated using Q-switched (nanosecond-pulsed) spectral components [1]. In this article, we extend this idea to the continuous-wave (CW) domain and demonstrate CW light modulation at a frequency of 90 THz. The modulation frequency in our experiment is about one third of the optical carrier frequency.

Our experiment builds on the pioneering work of Carlsten and colleagues, who were the first to demonstrate stimulated Raman scattering in gases with CW laser beams [2–4]. In stimulated Raman scattering, molecules pumped with sufficiently intense light at frequency ω_p produce Stokes light at $\omega_s = \omega_p - \omega_v$, where ω_v is the selected vibrational or rotational frequency of the molecule. If the molecules are placed inside a cavity with a high finesse at the pump and at the Stokes wavelengths, efficient Stokes generation in molecular gases can be achieved with CW laser beams. Advances in high-reflectivity, ultralow-loss dielectric coatings have allowed efficient Stokes generation with pump laser powers as low as 1 mW. Both rotational and vibrational CW Stokes sideband generation has been reported. The key difference of our work when compared with earlier experiments is that we drive a sufficiently large molecular coherence such that we not only generate the Stokes beam but also produce substantial anti-Stokes light at a frequency of $\omega_a = \omega_p + \omega_v$. The anti-Stokes beam is produced by a single pass through the system: that is, the cavity does not have a high finesse at the anti-Stokes wavelength. Specifically, by using the fundamental vibrational transition in molecular deuterium (D_2), we produce a CW spectrum with three discrete components at wavelengths 1.56 μm, 1.06 μm, and 807 nm. These wavelengths span 0.95 octaves of optical bandwidth. We then show that these beams have almost perfect mutual-phase coherence and thereby infer the synthesis of a near single-cycle optical wave form.

Our work has largely been motivated by the technique of adiabatic Raman generation at maximum coherence, which has been pioneered by Harris and Sokolov [5,6]. As mentioned previously, about nine years ago, this technique allowed the first light modulation at molecular frequencies. Building on this work, Shverdin *et al.* and Kung and colleagues have synthesized the first single-cycle optical pulses [7,8]. Recently, the synthesis of arbitrary optical wave forms using a broad Raman spectrum has also been demonstrated [9]. We view our experiment as the first step toward extending these pioneering efforts to the CW domain. In the future, by generating more than three spectral components, our technique may allow arbitrary wave-form synthesis with CW laser beams. In other related prior work, the use of stimulated Raman scattering for ultrashort-pulse synthesis was originally suggested by Imasaka and colleagues [10] and by Kaplan [11]. Hakuta and colleagues have produced a broad Raman spectrum by using solid molecular hydrogen [12]. Katsuragawa and coworkers have demonstrated an octave-spanning Raman comb with carrier-envelope phase control [13]. Utilizing an impulsive Raman excitation technique, Korn and colleagues have produced optical pulses as short as 3.8 fs [14]. Over the past decade, there has also been significant progress on CW-stimulated Raman scattering. Zaitsu *et al.* have demonstrated efficient CW rotational Raman generation at the anti-Stokes wavelength by tuning the cavity resonances with the help of a dispersive gas [15]. The same group has also demonstrated the generation of a second rotational Stokes beam through cascade-stimulated Raman scattering [16]. Couny and coworkers have demonstrated CW rotational Stokes output power of more than 2 W inside a hollow photonic crystal fiber [17]. We have recently demonstrated high-power CW rotational Raman generation at low gas pressures in molecular D_2 [18].

We proceed with a detailed description of our experiment. As shown in Fig. 1, the experiment is performed at room temperature inside a cavity with a high finesse at the pump (1.064-μm) and the Stokes (1.56-μm) wavelengths. The high-finesse cavity (HFC) is placed inside a vacuum chamber that we fill with D_2. The mirrors of the cavity have ultralow-loss, high-reflectivity dielectric coatings with a CW optical damage threshold exceeding 20 MW/cm^2. The total scattering, absorption, and transmission losses of the mirrors at the two wavelengths are at the level of 100 parts per million (ppm). The mirrors have a radius of curvature of 50 cm, and the length of the cavity is 27 cm. The free spectral range of the cavity is 555 MHz. The calculated finesse of the cavity at the two wavelengths is about 22,000 resulting in a full width at half maximum (FWHM) cavity resonance linewidth of 25 kHz. One of the mirrors is mounted on a piezo-electric

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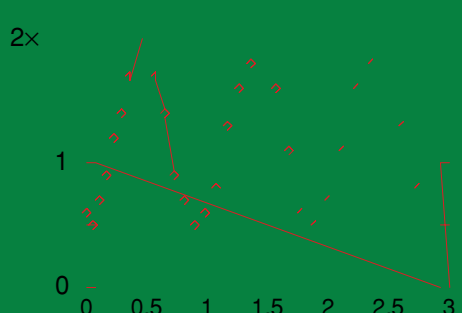
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the cavity and (2) phase control and therefore a primitive form of wave-form synthesis in CW-stimulated Raman scattering. We view our experiment as the first step toward synthesizing arbitrary optical waveforms using CW spectral components. We note that a key limitation of our experiment is the optical power of the generated spectrum. Due to the low generated anti-Stokes power, the synthesized wave form of Fig. 5 has an average optical power of only a fraction of a milliwatt. Cooling the D_2 molecules to their rotational ground state (by using liquid nitrogen, for example) is predicted to increase the generated optical power by at least an order of magnitude [6]. Other future investigations will include operating near maximum coherence [22] or using a multiplicative technique [23] to increase the generated number of Raman beams and thereby increase the number of degrees of freedom in temporal wave-form synthesis.

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