

Broadband spectrum generation using continuous-wave Raman scattering

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We experimentally demonstrate a continuous-wave (CW) ro-vibrational Raman spectrum that is two octaves wide with spectral components ranging from $0.8\ \mu\text{m}$ to $3.2\ \mu\text{m}$ in wavelength. The spectrum is produced in low-pressure molecular deuterium inside a high-finesse cavity.

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Since the invention of the laser in 1960, optical scientists have sought to develop light sources capable of generating coherent optical spectra covering the infrared, visible, and ultraviolet spectral regions. Broadband sources are of particular interest because they have applications in a wide range of research areas including ultrafast physics, precision spectroscopy, and quantum control [1–3]. With the invention of and the developments in Ti:sapphire-based light sources, spectral widths of 100 THz (corresponding to a transform limited pulse-width of ≈ 10 fs) are routinely achieved. A current important challenge in optical science is to develop reliable sources with a spectral width exceeding 1 petahertz (PHz), which would enable the synthesis of sub-femtosecond, sub-cycle optical pulses. A PHz wide optical source would also allow the construction of an arbitrary optical waveform generator, a device similar in functionality to an electronic radio-frequency synthesizer, but at optical frequencies [4].

Over the last two decades, there has been continuous progress towards constructing a PHz-wide optical source. Recently the generation of multi-octave broadband spectra has been demonstrated using a number of approaches, including coherently combining the supercontinuum output from multiple highly nonlinear fibers and using multiple optical parametric amplifiers seeded by a common laser source [5–8]. Another approach that shows considerable promise for the construction of a PHz source is the technique of molecular modulation [9–12]. In this technique, molecules are driven to a highly coherent state such that they become efficient frequency modulators. As a result, many vibrational or rotational Stokes and anti-Stokes sidebands can be generated, producing a broad spectrum covering the full optical region. The molecular modulation technique was the first to break the single-cycle barrier [13], and the Kung group recently used five vibrational sidebands produced in molecular hydrogen to synthesize nonsinusoidal optical waveforms [14].

All of the molecular modulation experiments described

in the previous paragraph were performed using Q-switched pulsed lasers. The duty cycle of pulsed lasers is low, less than 1 part in 10^7 , which severely restricts data rates. Furthermore, each sideband has a minimum linewidth that is determined by the duration of the Q-switched pulse, $1/(10\ \text{ns}) \approx 100$ MHz, which precludes their use in precision spectroscopic applications. In this letter, we report an experiment that we see as the first step in extending the molecular modulation technique into the CW domain. By using low-pressure molecular deuterium inside a high-finesse cavity, we produce a broad CW ro-vibrational spectrum that is two octaves wide, ranging from $0.8\ \mu\text{m}$ to $3.2\ \mu\text{m}$ in wavelength. To our knowledge, this is the broadest spectrum ever produced in the CW domain. We have also measured the absolute linewidth of the spectral components and found them to be at the 10 kHz level.

The details of our experimental set-up can be found in our previous publication [15]. Briefly, to produce a high power CW pump beam, we start with a custom-built external cavity diode laser (ECDL) with an optical power of 20 mW and a free-running linewidth of about 0.5 MHz. We amplify the ECDL output with a ytterbium-doped fiber amplifier centered at $1.064\ \mu\text{m}$. The amplifier produces a CW output power of 20 W in a single spatial mode. The amplified beam is then coupled to the TEM₀₀ mode of a high-finesse cavity using a mode matching lens. The mirrors of the high-finesse cavity have high damage threshold coatings with high reflectivity near wavelengths $1.06\ \mu\text{m}$ (pump) and $1.55\ \mu\text{m}$ (vibrational Stokes). The transmittance of the mirrors at the two wavelengths are about 50 parts-per-million (ppm), and the total scattering and absorption losses are at the level of 100 ppm. The calculated finesse of the cavity at the two wavelengths is about 20,000, resulting in a full-width-half-maximum cavity resonance linewidth of 10 kHz. One of the cavity mirrors is mounted on a piezoelectric transducer to allow for slight adjustments of the cavity length. We use the Pound-Drever-Hall technique to lock the high-power pump beam to the cavity. The

high finesse cavity is housed in a stainless-steel vacuum chamber whose central 50 cm-long region is surrounded by a liquid-N₂ reservoir. Cooling the molecules reduces Doppler broadening and also greatly increases the population of the ground rotational level.

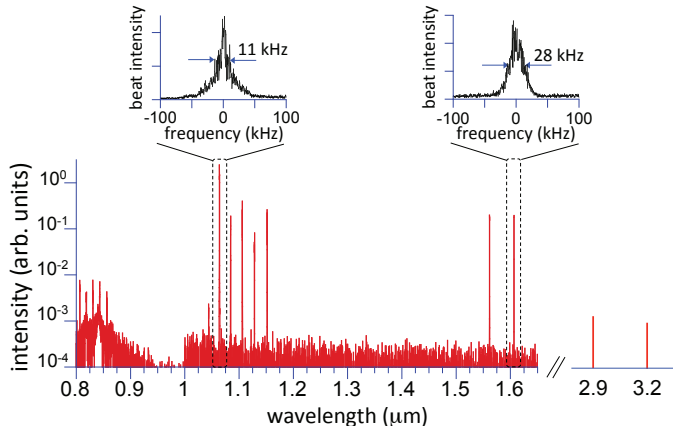


Fig. 1. (Color online) The ro-vibrational spectrum generated in molecular D₂ using a cavity with a high finesse near wavelengths 1.06 μm and 1.55 μm . The spectrum is obtained by locking the 1.06 μm pump laser to the cavity. The 0.8-1.65 μm region is observed on an optical spectrum analyzer in a single scan, and the two-mid-infrared sidebands (2.9 μm and 3.2 μm) are measured using a PbSe photo-diode. In the spectrum analyzer scan, the change in the background level between 0.8-0.9 μm and the jump in the background level at 1 μm are instrument artifacts. The insets show absolute linewidth measurements at the 1.06 μm and 1.6 μm sidebands.

At molecular gas pressures of order 0.1 atm, we observe that the vibrational Stokes sideband at 1.55 μm on the $|\nu = 0, J = 0\rangle \rightarrow |\nu = 1, J = 0\rangle$ transition is generated through Raman lasing when the pump laser at 1.06 μm is locked to the cavity [16]. The pump and the generated Stokes beams drive the coherence and produce the vibrational anti-Stokes (0.8 μm) and second Stokes (2.9 μm) sidebands through four-wave mixing. The mirrors do not have high reflectivity near 0.8 μm nor in the mid-infrared. As a result, the vibrational anti-Stokes and second Stokes sidebands are generated in a single pass through the cavity. In addition to vibrational Raman generation, we observe rotational Raman lasing on the $|\nu = 0, J = 0\rangle \rightarrow |\nu = 0, J = 2\rangle$ transition. This is because the mirrors of the cavity have sufficiently high reflectivity in the wavelength ranges 1.0-1.18 μm and 1.50-1.65 μm that this rotational generation builds up from noise. Furthermore, the generated rotational Stokes beam can behave as an independent pump and produce the subsequent higher-order Stokes sideband, again through Raman lasing. As a result, we produce multiple-order rotational Stokes sidebands through cascade Raman lasing. Since this process happens in addition to vibrational Raman generation, we observe a broad ro-vibrational spectrum with frequencies of the form $\nu_{\text{pump}} + q\nu_M^{\text{vib}} + r\nu_M^{\text{rot}}$ ($q, r = \text{integers}$). $\nu_M^{\text{vib}} = 90 \text{ THz}$ ($|\nu = 0, J = 0\rangle \rightarrow |\nu = 1, J = 0\rangle$ vibrational transition),

and $\nu_M^{\text{rot}} = 5.3 \text{ THz}$ ($|\nu = 0, J = 0\rangle \rightarrow |\nu = 0, J = 2\rangle$ rotational transition). Figure 1 shows the generated spectrum that is transmitted through one of the cavity mirrors. The 0.8-1.65 μm region is observed on an optical spectrum analyzer in a single scan. The combined power of the two mid-infrared sidebands (2.9 μm and 3.2 μm) is measured using a PbSe photo-diode. The measured optical powers in the pump (near 1.06 μm), vibrational Stokes (near 1.55 μm), vibrational anti-Stokes (near 0.8 μm) and vibrational second Stokes (near 2.9 μm) regions are 15 mW, 20 mW, 50 μW , and 1 μW respectively. The optical power in the individual rotational sidebands in each region can be estimated using the vertical scale of the spectrum analyzer scan. These power values can be tuned somewhat since we can typically adjust the relative strength of vibrational and rotational generation by slightly changing the cavity length. The Raman lasing process depends critically on the overlap of the position of the longitudinal cavity modes with the Raman transition frequencies. Changing the cavity length tunes the position of the cavity modes in or out of resonance with a particular Raman transition, thereby adjusting its strength.

We have performed an absolute linewidth measurement of the spectral components in the pump and the vibrational Stokes regions of the spectrum. For this purpose, we have built an optical heterodyne detector. The detector splits a sideband into two paths and delays one arm using a 10 km-long fiber (which corresponds to an optical delay time of about 50 μs). We then measure the beat intensity as a function of frequency by combining the two arms on a fast photo-diode. We have found the beat linewidth measurements to be sensitive to our experimental conditions, such as the quality of the Pound-Drever-Hall lock. As a result, the linewidth measurements can vary by as much as a factor of two from day to day. However, for each of the seven spectral components we have examined (five near the pump and two near the vibrational Stokes), we have found the beat linewidth to be consistently at the 10 kHz level. As an example, the insets in Fig. 1 show the beat measurements at 1.06 μm and 1.6 μm taken at a particular day. The full-width-half-maximum linewidths at the two wavelengths are found to be 11 kHz and 28 kHz respectively. We believe that the absolute linewidths of the sidebands are currently limited by the imperfections in the locking electronics and also the mechanical perturbations to the cavity. It is likely that the absolute linewidths can be made smaller than 1 kHz with technical improvements in our setup [16].

Figure 2 shows the spatial profiles of the ten ro-vibrational sidebands near the pump (1.06 μm) and vibrational anti-Stokes (0.8 μm) regions of the spectrum. In this measurement, the generated spectrum transmitted through one of the cavity mirrors is spatially dispersed with an SF10 prism. The beams are then captured using a CCD-based beam profiler. All the beams have near perfect Gaussian spatial profiles since the beams

are produced in a TEM₀₀ mode of the high finesse cavity. We have found that by changing the cavity length and thereby tuning the position of the cavity resonances, we can cause some of the ro-vibrational sidebands to lase in higher order spatial modes of the cavity. For this case, we typically observe reduced efficiency in ro-vibrational sideband generation. A detailed study of ro-vibrational spectra generation using higher-order modes of the cavity will be among our future investigations.

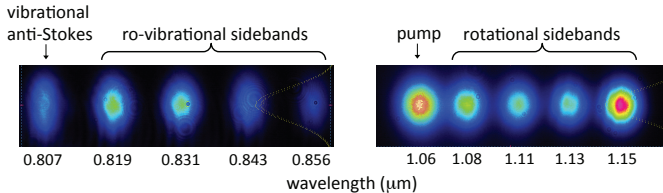


Fig. 2. (Color online) A CCD image displaying the spatial profiles of the ro-vibrational sidebands near the pump (1.06 μm) and the vibrational anti-Stokes (0.8 μm) regions of the spectrum.

We have also observed simultaneous rotational generation on multiple transitions in addition to vibrational generation. At liquid-N₂ temperature, the thermal populations of the $J = 0$, $J = 1$, and $J = 2$ rotational levels are 46%, 45%, and 8%, respectively. As a result, if the cavity is appropriately tuned, rotational scattering starting from each of these J levels is possible. For example, Fig. 3 highlights the details of the pump and vibrational anti-Stokes regions of the spectrum at a pressure of 0.2 atm. Here the pump produces five rotational sidebands on the $|\nu = 0, J = 0\rangle \rightarrow |\nu = 0, J = 2\rangle$ transition (AS1 through S4). In addition, a rotational Stokes sideband on the $|\nu = 0, J = 1\rangle \rightarrow |\nu = 0, J = 3\rangle$ transition ($\nu_M = 8.9$ THz) is produced (P'). This beam then generates the primed spectrum (AS1' through S2') by scattering on the $|\nu = 0, J = 0\rangle \rightarrow |\nu = 0, J = 2\rangle$ excitation. Another separate Stokes sideband from the pump beam is generated on the $|\nu = 0, J = 2\rangle \rightarrow |\nu = 0, J = 4\rangle$ transition ($\nu_M = 12.4$ THz), which similarly produces the double-primed spectrum on the $|\nu = 0, J = 0\rangle \rightarrow |\nu = 0, J = 2\rangle$ transition (AS2'' through S2''). All the rotational anti-Stokes sidebands and the vibrational anti-Stokes region of the spectrum are produced through four-wave mixing. Including the sidebands near 1.55 μm and 2.9 μm (not shown in Fig. 3), this approach produces about 30 ro-vibrational sidebands.

We next discuss the applications of our approach to temporal waveform synthesis. Because there is not a rational relationship between the pump frequency and vibrational and rotational frequencies ($\nu_M^{vib} = 90$ THz and $\nu_M^{rot} = 5.3$ THz), the synthesized waveform is quasi-periodic. For example, if we assume that all the spectral components are adjusted to have equal amplitudes and phases, the spectrum of Fig. 4 would synthesize a quasi-periodic train of pulses with a large pulse-to-pulse carrier-envelope phase-drift. Some of these pulses in the pulse train would have a sub-cycle cosine shape with a duration of 0.9 fs. Synthesizing such waveforms will re-

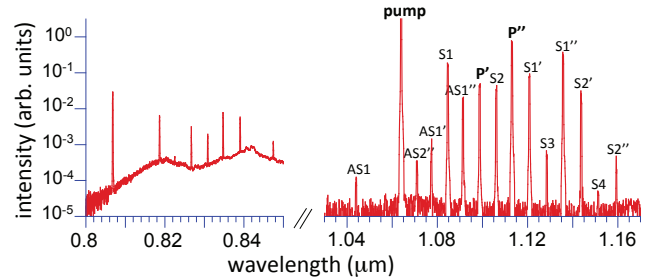


Fig. 3. (Color online) Generated spectra using a vibrational and three rotational excitations. The rise in the background level between 0.8-0.85 μm is an instrument artifact.

quire phase and amplitude control of the generated spectrum which will be one of our major goals in the near future. The temporal waveforms can be measured using cross-correlation studies, similar to the ones that have been performed with the recent pulsed molecular modulation experiments [13, 14].

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