

# High-frequency modulation of continuous-wave laser beams by maximally coherent molecules

D. D. Yavuz

*Department of Physics, University of Wisconsin, 1150 University Avenue, Madison, Wisconsin 53706, USA*

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We suggest a technique where a molecular Raman transition is prepared near a maximally coherent state with continuous-wave (CW) laser beams. The key idea is to put the molecules inside a high-finesse cavity and to drive the molecular transition with two lasers whose frequency difference is slightly detuned from the Raman resonance. In this regime, the molecules become very efficient CW modulators with a modulation frequency that can be as high as 100 THz.

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Over the last decade, there has been a growing emphasis on the use of coherent Raman scattering for broadband spectrum generation and ultrashort pulse synthesis. Since the original suggestions by Yoshikawa and Imasaka [1] and Kaplan [2], a number of experiments have demonstrated generation of ultrashort pulses using broadband Raman spectra. Utilizing an impulsive excitation technique, Zhaborov and Korn have produced pulses as short as 3.8 fs [3]. Recently, using an adiabatic Raman excitation technique, Shverdin *et al.* at Stanford have demonstrated the synthesis of the first single-cycle optical pulse [4].

The adiabatic excitation technique of Shverdin *et al.*, termed “molecular modulation,” relies on broadband Raman generation in the regime of maximum coherence [5,6]. Here, the idea is to drive a molecular transition with two single-mode, linearly polarized laser fields whose frequency difference is slightly detuned from the frequency of the Raman resonance. Note in Fig. 1, when the detuning from the Raman transition is sufficiently large, the excitation is adiabatic, and population smoothly flows from the ground state  $|a\rangle$  to excited state  $|b\rangle$ . For sufficiently intense driving laser beams, it becomes possible to transfer almost half of the population and to approach the state of maximum coherence. When this is the case, the magnitude of the coherence of the Raman transition (the off-diagonal density matrix element) is near its maximum value,  $|\rho_{ab}| \approx 1/2$ . The established molecular coherence mixes with the driving lasers and a broad Raman spectrum is generated collinearly with phase matching playing a negligible role. In essence, molecules act as frequency modulators with a modulation frequency that can be as high as 100 terahertz (THz). Using the molecular modulation technique in molecular deuterium ( $D_2$ ), Sokolov *et al.* demonstrated generation of 17 vibrational sidebands ranging from 195 nm to 2.94  $\mu$ m in wavelength [7]. Yavuz *et al.* demonstrated generation of 200 rovibrational sidebands by simultaneously exciting a rotational and a vibrational transition [8]. Hakuta and co-workers extended this technique to Raman transitions in molecular solid hydrogen ( $H_2$ ) [9,10].

Molecular modulation technique as described in the previous paragraph can also be used as an efficient single sideband generator. Here, the idea is to drive the Raman transition with two circularly polarized laser fields. When this is the case, angular momentum conservation prohibits the generation of additional sidebands from the driving lasers. However, a molecular local oscillator is prepared which can act as

a frequency mixer. A third laser beam with appropriate polarization can then be frequency shifted producing a single sideband spaced by the Raman excitation frequency. Using this technique, Sokolov *et al.* demonstrated a 15% efficient single sideband converter at a frequency spacing of 95 THz [11].

All of the molecular modulation experiments discussed in the previous paragraphs have been done using *Q*-switched, high power, pulsed lasers. As a result, each sideband has a minimum linewidth determined by the duration of the *Q*-switched pulse,  $1/(10 \text{ ns}) \approx 100 \text{ MHz}$ . For precision spectroscopy applications, the extension of the molecular modulation technique to continuous-wave (CW) lasers would be extremely important. Also, *Q*-switched lasers suffer from many limitations such as low repetition rate and pulse-to-pulse intensity and timing jitter which would be absent for CW lasers. In this paper we demonstrate a scheme where single sideband conversion using molecular modulation is accomplished with CW lasers. The key idea is to put the molecules inside a cavity that has a high finesse at the two driving laser beams. We predict that, using parameters within

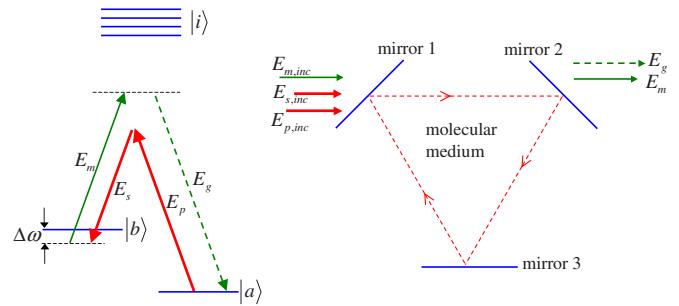


FIG. 1. (Color online) Energy level diagram and the schematic of the suggested technique. Two CW, opposite circularly polarized laser beams,  $E_p$  and  $E_s$ , drive  $|a\rangle$  to  $|b\rangle$  Raman transition. When the magnitude of the coherence of the Raman transition approaches its maximum value,  $|\rho_{ab}| \approx 1/2$ , molecules act as an efficient frequency mixer. A third laser beam with appropriate polarization,  $E_m$ , can then be frequency shifted to produce  $E_g$ . To obtain the high intensities required for the preparation of near-maximum coherence with CW lasers, we utilize a ring cavity that has a high finesse at the two driving laser beams. The quantities  $E_{p,\text{inc}}$ ,  $E_{s,\text{inc}}$ , and  $E_{m,\text{inc}}$  denote the field values of the laser beams that are incident on the cavity. The third laser,  $E_m$ , does not need to be resonant with the cavity and can be arbitrarily weak.

the reach of current experiments, one can prepare a near-maximum molecular coherence with two CW lasers. As a result, a CW local oscillator at the molecular transition frequency is prepared. A third, widely tunable laser beam, that does not need to be resonant with the cavity, can then be frequency shifted by the frequency of the molecular oscillation.

Our scheme has been motivated by the recent pioneering experiments of Carlsten and co-workers that have achieved CW Raman lasing inside a high finesse cavity [12–15]. In related work, Shinzen *et al.* have investigated the temporal coherence of a CW Raman laser beam [16]. Ihara *et al.* have discussed multiple order Raman scattering inside a high finesse cavity [17]. The key difference of our approach when compared to these earlier experiments is this: once maximum molecular coherence is prepared, our system can modulate any other laser that can be widely tuned and that does not need to be resonant with the cavity. In contrast, in earlier approaches, laser light is generated only at wavelengths where the cavity has a very high finesse. We also note that there is substantial literature on four-wave mixing utilizing double  $\Lambda$  schemes in atomic systems [18]. The use of molecular systems such as molecular H<sub>2</sub> has the following key advantages over the atomic systems: (1) With molecules, it is easy to obtain large density length products at room temperature. This removes the need for complicated heat ovens. (2) Due to the large detuning from the electronic states, the molecular systems are, in principle, widely tunable and may be used as frequency converters in a wavelength region ranging from  $\lambda=100$  nm to  $\lambda=10 \mu\text{m}$ .

We proceed with a detailed description of our scheme. We first describe the preparation of the molecular coherence with the two driving laser beams (termed the pump laser,  $E_p$ , the Stokes laser,  $E_s$ ). Note in Fig. 1 the beams are incident to a cavity that has a high finesse at the frequencies of the two driving lasers. To avoid complications due to the interference of counterpropagating beams, we consider a ring cavity instead of a standing-wave cavity. We assume the cavity to be filled with molecular medium of interest, such as molecular H<sub>2</sub>. Figure 1 shows the energy level diagram for a typical molecular system. Here state  $|a\rangle$  is the ground rovibrational state of the molecule. State  $|b\rangle$  is a particular excited rovibrational state, and states  $|i\rangle$  are high lying excited electronic rovibrational states. In light diatomic molecules, the energy spacing between the rovibrational states is large enough such that the molecules can be initially prepared in the ground state  $|a\rangle$  by cooling with liquid nitrogen. The two photon detuning of the driving laser beams from the Raman resonance is defined as  $\Delta\omega=(\omega_b-\omega_a)-(\omega_p-\omega_s)$ . To avoid dissipation and to prepare a single molecular eigenstate, we consider the case where the two photon detuning is much larger than the dephasing linewidth of the Raman excitation,  $\Delta\omega\gg\gamma$ . In this regime, starting at the ground state  $|a\rangle$ , the molecular excitation adiabatically follows the driving laser beams. Throughout this paper, we will take the mixing beam  $E_m$  to be much weaker when compared with the two driving lasers  $E_p$  and  $E_s$ . We, therefore, ignore the effect of the mixing beam on the molecular states. We define  $B=|B|\exp(j\varphi)$  and  $\tan\theta=2|B|/(2\Delta\omega-D+A)$ . Here  $B$  is the two photon drive for the Raman transition,  $B=bE_pE_s^*$ , and the quantities

$A$  and  $D$  determine the Stark shift of the two Raman states  $A=a_p|E_p|^2+a_s|E_s|^2$ ,  $D=d_p|E_p|^2+d_s|E_s|^2$ . The constants  $a$ ,  $b$ , and  $d$  are summations which include dipole matrix elements  $\mu_{ai}$  and  $\mu_{bi}$  and also the detuning of the laser beams from the excited states  $|i\rangle$ . With these definitions, in the presence of the laser beams, the density matrix elements of the Raman transition are [5,6]

$$\rho_{aa}=\cos^2(\theta/2), \quad \rho_{bb}=\sin^2(\theta/2),$$

$$\rho_{ab}=\left(\frac{1}{2}\sin\theta\right)e^{j\varphi}. \quad (1)$$

Inside the cavity, the following slowly varying envelope Maxwell's equations describe the propagation of the two beams [5,6]:

$$\frac{\partial E_p}{\partial z}=-j\eta\hbar\omega_pN(a_p\rho_{aa}E_p+d_p\rho_{bb}E_p+b^*\rho_{ab}E_s),$$

$$\frac{\partial E_s}{\partial z}=-j\eta\hbar\omega_sN(a_s\rho_{aa}E_s+d_s\rho_{bb}E_s+b\rho_{ab}^*E_p), \quad (2)$$

where  $N$  is the molecular density and  $\eta=(\mu/\epsilon_0)^{1/2}$ . The two beams are coupled to each other through the molecular coherence,  $\rho_{ab}$ . Equation (2) assumes that the beams are large in the transverse dimension and neglects diffractive effects. Equations (1) and (2) describe the evolution of the molecular system and the propagation of the two laser beams. In addition, we need to consider the effect of the mirrors on the laser beams. For simplicity, we will take all three mirrors to be identical, and the reflection and transmission coefficients of the mirrors for both laser beams to be the same. Denoting the amplitude reflection and transmission coefficients of the mirrors with  $r$  and  $t$ , the two-port equation for the two laser beams at the first mirror is [19]

$$E_p(0)=jtE_{p,\text{inc}}+rE_p(L),$$

$$E_s(0)=jtE_{s,\text{inc}}+rE_s(L). \quad (3)$$

Here  $L$  is the length of the cavity and  $E_{p,\text{inc}}$  and  $E_{s,\text{inc}}$  denote the field values of the two laser beams incident on the cavity. For simplicity we take all three mirrors to be equally spaced along the length of the cavity. The two-port equations at the other two mirrors are  $E_p(L/3^+)=rE_p(L/3^-)$ ,  $E_s(L/3^+)=rE_s(L/3^-)$  for mirror 2 and  $E_p(2L/3^+)=rE_p(2L/3^-)$ ,  $E_s(2L/3^+)=rE_s(2L/3^-)$  for mirror 3, respectively. Here, “−” denotes the field value right before it hits the mirror and “+” denotes the field value right after it hits the mirror, respectively.

We now proceed with a self-consistent and stable steady-state solution for the density matrix elements [Eq. (1)] and for the two driving laser beams inside the cavity [Eqs. (2) and (3)]. It can be shown that, between the first and the second mirrors,  $0 < z < L/3$ , the solutions for the electric-field amplitudes of the two beams and the density matrix elements of the Raman transition are

$$E_p(z)=E_p(0)\exp(-j\phi_{p,1}z),$$

$$E_s(z)=E_s(0)\exp(-j\phi_{s,1}z),$$

$$\begin{aligned}\rho_{ab}(z) &= \rho_{ab}(0)\exp[-j(\phi_{p,1} - \phi_{s,1})z], \\ \rho_{aa}(z) &= \rho_{aa}(0), \quad \rho_{bb}(z) = \rho_{bb}(0).\end{aligned}\quad (4)$$

The quantities  $\phi_{p,1}$  and  $\phi_{s,1}$  are the propagation constants that determine the phase accumulation of the two beams and they are

$$\phi_{p,1} = \eta\hbar\omega_p N \left( a_p \rho_{aa}(0) + d_p \rho_{bb}(0) + b^* \rho_{ab}(0) \frac{E_s(0)}{E_p(0)} \right),$$

$$\phi_{s,1} = \eta\hbar\omega_s N \left( a_s \rho_{aa}(0) + d_s \rho_{bb}(0) + b \rho_{ab}^*(0) \frac{E_p(0)}{E_s(0)} \right). \quad (5)$$

Similar solutions can be found for the laser beam amplitudes and density matrix elements in other regions of the cavity. We denote the propagation constants of the two lasers between mirrors 2 and 3 with  $\phi_{p,2}$  and  $\phi_{s,2}$ , and between mirrors 3 and 1 with  $\phi_{p,3}$  and  $\phi_{s,3}$ , respectively. With these definitions, using the two-port equation at the first mirror [Eq. (3)], we can find the amplitude of the circulating fields inside the cavity

$$\begin{aligned}E_p(0) &= \frac{jt}{1 - r^3 \exp[-j\omega_p L/c - j(\phi_{p,1} + \phi_{p,2} + \phi_{p,3})L/3]} E_{p,\text{inc}}, \\ E_s(0) &= \frac{jt}{1 - r^3 \exp[-j\omega_s L/c - j(\phi_{s,1} + \phi_{s,2} + \phi_{s,3})L/3]} E_{s,\text{inc}}.\end{aligned}\quad (6)$$

If the total phase accumulation while propagating through the cavity is a multiple of  $2\pi$ , then the cavity will be resonant with the field and high intensity will buildup. From Eq. (6) the resonance condition for both beams is achieved when  $\omega_p L/c + (\phi_{p,1} + \phi_{p,2} + \phi_{p,3})L/3 = q_1(2\pi)$  and  $\omega_s L/c + (\phi_{s,1} + \phi_{s,2} + \phi_{s,3})L/3 = q_2(2\pi)$ , where  $q_1$  and  $q_2$  are integers. These two resonant conditions can always be satisfied by tuning the cavity length  $L$ , and by also simultaneously tuning the frequencies of the driving lasers  $\omega_p$  and  $\omega_s$ , while keeping their frequency difference constant. When both of the resonance conditions are satisfied, the cavity fields will buildup to a large value given by  $E_p(0) = \frac{jt}{1-r^3} E_{p,\text{inc}}$  and  $E_s(0) = \frac{jt}{1-r^3} E_{s,\text{inc}}$ . The large intensities in the cavity strongly drive the Raman transition as per Eq. (1). We then have a self-consistent solution for the propagation of the two fields inside the cavity and also for the density matrix elements of the molecular transition as described through Eqs. (1), (4), and (5).

We next describe the modulation process of a third, weak laser beam. Once the molecules are driven to a state of large molecular coherence, any other laser beam can beat with the established coherence and produce a sideband frequency shifted by the frequency of the molecular Raman transition. For simplicity, we assume that the mirrors are completely transmissive at the mixing and generated wavelengths and therefore ignore the cavity feedback for these fields. Denoting the amplitude of the generated field to be  $E_g$ , these fields evolve according to

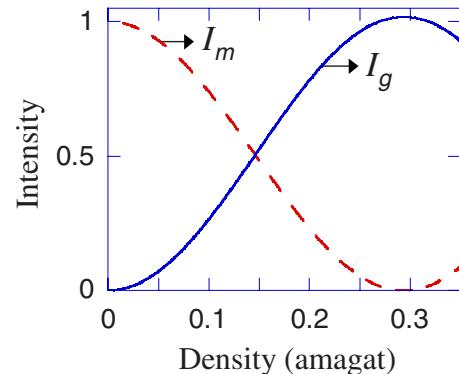


FIG. 2. (Color online) The intensities of the mixing beam,  $I_m = |E_m|^2/2\eta$ , and the generated beam,  $I_g = |E_g|^2/2\eta$ , at  $z=L/3=10$  cm as a function of density of the molecular H<sub>2</sub> (1 amagat corresponds to 1 atmosphere pressure at room temperature). At a density of about 0.15 amagats, 50% of the power in the mixing beam is converted into the generated beam and the system acts as an amplitude modulator at a modulation frequency of 10.62 THz. At a density of about 0.3 amagats, all of the energy in the mixing beam is converted into the generated beam and the system acts as a 100% efficient frequency up converter

$$\frac{\partial E_g}{\partial z} = -j\eta\hbar\omega_g N(a_g \rho_{aa} E_g + d_g \rho_{bb} E_g + b_g^* \rho_{ab} E_m),$$

$$\frac{\partial E_m}{\partial z} = -j\eta\hbar\omega_m N(a_m \rho_{aa} E_m + d_m \rho_{bb} E_m + b_g \rho_{ab}^* E_g). \quad (7)$$

We proceed with a numerical example. We take our molecular medium to be molecular H<sub>2</sub> and consider  $S_0(0)$  rotational transition. The two Raman states are  $|a\rangle \equiv |\nu=0, J=0, m_J=0\rangle$  and  $|b\rangle \equiv |\nu=0, J=2, m_J=2\rangle$  ( $\nu$  is the vibrational quantum number and  $J$  is the rotational quantum number). The frequency of the Raman excitation is  $\omega_b - \omega_a = 354.35$  cm<sup>-1</sup> (10.62 THz). We take the two driving laser beams to be opposite circularly polarized and choose their wavelengths to be  $\lambda_p = 800$  nm and  $\lambda_s = 823.3$  nm. We take the two photon detuning from the Raman transition to be  $\Delta\omega = 2\pi \times 1$  GHz. While calculating the constants  $a$ ,  $b$ , and  $d$  of Eqs. (1), (2), and (7), we consider all allowed electronic rovibrational transitions in the Lyman and Werner bands of molecular H<sub>2</sub>. We obtain the transition frequencies and the oscillator strengths for these transitions from the work of Abgrall [20]. For the pump laser beam,  $E_p$ , we calculate these quantities to be  $a_p = 2.44 \times 10^{-7}$ ,  $b_p = 1.94 \times 10^{-7}$ , and  $d_p = 2.62 \times 10^{-7}$  in mks units.

We consider a high finesse cavity with parameters similar to the ones used by Carlsten and co-workers [12–15]. We take the power reflection coefficients for all mirrors to be  $R \equiv r^2 = 0.9995$ . We take the incident power on the cavity for each driving laser beam to be 100 W. We take the spacing between the mirrors to be  $L/3 = 10$  cm and assume a Gaussian mode radius of  $W_0 = 100$  μm. For these parameters, when the resonance condition is satisfied, the circulating power for both laser beams inside the cavity is 88.9 kW. The circulating intensity inside the cavity is 566 MW/cm<sup>2</sup>. These power and intensity values are similar to those that were recently

achieved by Meng *et al.* [21]. For these values, the magnitude of the coherence of the molecular transition is  $|\rho_{ab}|=0.066$  and is maintained through the cavity.

Once the molecular coherence is prepared, it serves as a local oscillator and it can mix with a mixing field  $E_m$  to generate a single sideband  $E_g$  frequency upshifted by the frequency of the Raman excitation. We take the wavelength of the mixing beam to be  $\lambda_m=500$  nm which results in  $\lambda_g=491$  nm for the generated beam. With the density matrix elements for the molecular system calculated by Eq. (4), we numerically solve Eq. (7) with the boundary condition that  $E_m(0)=E_{m,\text{inc}}$  and  $E_g(0)=0$ . In Fig. 2 we plot the intensities of the mixing beam  $I_m=|E_m|^2/2\eta$  and the generated beam  $I_g=|E_g|^2/2\eta$  at  $z=L/3=10$  cm as a function of density of the molecular H<sub>2</sub>. At a density of 0.15 amagats (1 amagat corresponds to 1 atmosphere pressure at room temperature), the conversion efficiency from the mixing beam to the generated beam is 50% and the system acts as an amplitude modulator at a modulation frequency of 10.62 THz. At a density of about 0.3 amagat, all of the energy in the mixing beam is converted into the generated beam. The system therefore acts as a 100% efficient frequency up converter.

We next discuss how the conversion efficiency from the mixing beam to the generated beam scales when we vary the parameters of our scheme. The conversion efficiency is high-

est for a state of maximum molecular coherence,  $|\rho_{ab}|=1/2$ . Before this maximum limit is achieved, the conversion efficiency scales as  $\propto I_p I_s N^2 L^2$ . Here,  $I_p$  and  $I_s$  are the intensities of the pump and the Stokes lasers inside the cavity, respectively. With all the other parameters fixed, if we assume 10 times less power for the pump and Stokes lasers in the numerical simulation of Fig. 2, the conversion efficiency would, therefore, be about 1%. We also note that, since the molecular states are prepared with the strong driving lasers, the equations that describe the mixing process [Eq. (7)] are linear in the field amplitudes  $E_m$  and  $E_g$ . As a result, the incident mixing beam,  $E_{m,\text{inc}}$ , can be arbitrarily weak and can be widely tuned, from about  $\lambda_m=100$  nm to  $\lambda_m=10 \mu\text{m}$ .

To conclude, we have suggested a scheme where molecular modulation is achieved with CW lasers. Although throughout this paper we have taken the mixing beam to be monochromatic, our scheme can also be used to modulate a broadband laser such as a Ti:Sapphire laser [22]. We expect that our scheme will find applications in precision spectroscopy and in ultrashort pulse synthesis using Raman sidebands.

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