Near Quantum-Limited Phase Memory in a Raman Amplifier

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We have detected molecular coherence lasting nearly 9 times the collisional dephasing time in a Raman amplifier. Two laser pulses, separated in time, pumped a common set of hydrogen molecules, producing two Stokes pulses whose relative phase was measured interferometrically. The variance of this phase is a direct measure of the memory of the molecular coherence induced by the first scattering process. This experiment is capable of detecting a level of molecular coherence equivalent to between 15 and 50 vibrational phonons in the most strongly excited mode.

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The level at which quantum noise dominates over coherent excitations is a fundamental question in physics. Several researchers have probed this question by examining the level of external optical input needed to influence the output field of a quantum noise amplifier. For example, Swanson, Battle, and Carlsten have recently found that it takes, on average, about five photons per (temporal) mode to control the phase of the output field of a Raman amplifier [1]. A similar result has been seen in optical parametric amplification [2]. Spatial mode control of Raman amplifiers and phase conjugators with coherent input has also been observed [3].

In contrast to using light to seed the amplification, we have investigated the level of initial molecular coherence needed to control the phase of the output field of a Raman amplifier. Two laser pulses, separated in time, were used to pump a common set of hydrogen molecules producing two Stokes pulses via stimulated Raman scattering (SRS). The relative phase of the two Stokes pulses was measured interferometrically. The variance of this relative phase is a direct measure of the memory of the molecular coherence that was induced by the first scattering process. Correlations between the two Stokes phases were observed even after nine molecular dephasing times, when the level of coherence had decayed by a factor of e^{-9}. We show that this extraordinarily long-lived phase memory comes about because a molecular coherence corresponding on average to fewer than 50 vibrational phonons is sufficient to influence the phase of the output field. Recently, Marshall and Piper have invoked this same long-lived molecular coherence to explain an observed gain enhancement when using multipeaked laser pulses to pump an SRS generator [4].

Under proper conditions, SRS produces a well collimated beam of light with relatively narrow bandwidth, a high degree of spatial coherence, and many photons per mode. With short pump pulses, yielding scattering in the transient regime, there exists a well-defined phase and amplitude during the amplification process. However, since the scattering process builds up from quantum noise, the resulting amplitude and phase are expected to be random from shot to shot [5]. We have shown previously that SRS produces light statistics that are thermal-like in the transient, unsaturated regime [6]. The fluctuation of the phase of the Stokes field was indeed observed to be uniform and randomly distributed. Departures from uniform and random phase statistics can be a sensitive probe of possible underlying correlations present between separate scattering events in a Raman amplifier. This is the fundamental principle behind our present experimental technique.

Figure 1 shows the experimental configuration. A Q-switched, cavity dumped, mode-locked Nd-doped yttrium aluminum garnet laser is frequency doubled to produce 300-ps pump pulses at 532 nm with a 10-Hz repetition rate. Each pump pulse in the laser beam is collimated to a diameter of 2 mm to minimize diffraction effects and is split into two equally intense pulses by a 50% beam splitter. One of the pair of pump pulses is delayed by a variable time τ with respect to the other. The two beams are then recombined and carefully overlapped spatially by a second 50% beam splitter in order to ensure that they pump the same set of molecules in a 1-m-long H_2 cell at 33 atm. Before entering the cell, the spatially combined pump beams are recollimated to a diameter of 250 μm

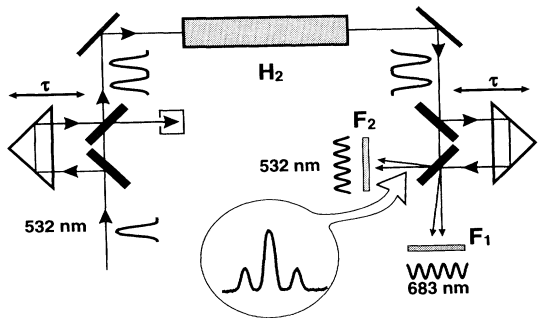

FIG. 1. Apparatus to measure the variance of the relative phase of two Stokes pulses at 683 nm, generated from a common set of H_2 molecules by two pump pulses (532 nm) separated by a time τ. The two Stokes pulses are temporally superposed and interfered to produce spatial fringes, allowing the relative phase to be determined.

over the length of the cell. This collimation ensures that the SRS process will excite only the lowest-order spatial mode and that a plane-wave description of the process is approximately valid [7]. Each pump pulse has a peak intensity I_p of $(3.0\pm0.3)\times10^8$ W/cm^2 at the entrance to the cell. The SRS scattering process occurs at 683 nm via the $Q(1)$ vibrational transition in H_2. The collisional dephasing rate Γ is 5.4×10^9 rad/sec at this pressure, while the steady-state Raman gain coefficient is $g_0=2.5\times10^{-9}$ cm/W [8]. The first, or leading, pump pulse produces Stokes light and a molecular coherence by SRS in the transient regime, $\Gamma\tau_p < g_0 I_p L$, where $\tau_p=300$ ps is the pump pulse duration (full width at half maximum) and L is the length of the gain medium [7]. The second, or trailing, pump pulse scatters off the remaining molecular coherence to produce a trailing Stokes pulse with some degree of phase correlation with respect to the leading Stokes pulse.

At the output of the cell there are two pump pulses and two Stokes pulses. Two 50% beam splitters and a second delay line allow us to temporally superimpose half of the leading-Stokes/pump pulse pair with half of the trailing-Stokes/pump pair. The inset in Fig. 1 shows a streak-camera trace of the resulting pump-pulse sequence. The central peak contains the superimposed pulses and thus gives rise to spatial fringes when the final 50% beam splitter is tilted at a slight angle. The side pulses, that is, the undelayed and twice-delayed pulses, give a background which limits the fringe visibility to a maximum of 50%. Spatial fringes due to the interference of the two Stokes pulses are produced in the same manner. By use of colored-glass filters and a charge-coupled-device camera, time-integrated interference patterns for both the pump and Stokes pulses are digitized and recorded.

On each shot the relative phase ϕ_n of the interfered Stokes pulses was extracted by fitting the interference pattern with a cosine function multiplied by a Gaussian envelope. At least 2000 shots were taken for each time delay. To compensate for slow thermal drift of the interferometer, we analyzed the data in terms of the difference of relative phases on successive shots, $\varphi_n=\phi_{n+1}-\phi_n$. The variance of this quantity was calculated for each time delay and is shown in Fig. 2. The variance of the phase is not zero for short delay times, as might be expected, because of the interferometer mechanical jitter. As a diagnostic of this jitter, the pump interference patterns were analyzed to obtain the average variance of the interferometer phase, shown as the dashed line in the figure. This measurement shows that the Stokes phase fluctuation at small delay is accounted for by the interferometer jitter. The average value of the interferometer phase variance is used as the error bars shown in Fig. 2 for the Stokes data. Repeated measurements of the Stokes phase variance at a few selected time delays show this to be a reasonable estimate. Correlations of the relative Stokes phases are seen for delays as long as 8.8 collisional dephasing times.

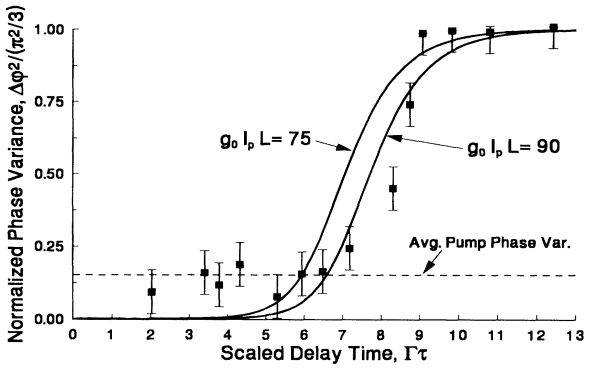

FIG. 2. Measurements (squares) and theoretical predictions (solid curves) of the phase variance vs time delay. The phase variance is normalized to the variance of a uniform, random phase. The time delay is scaled by the collisional dephasing rate Γ. The dashed line is the average variance of the pump-pulse fringes, used as a measure of the interferometer stability. The theoretical curves are labeled by the steady-state gain-length product, with $g_0 I_p L=75$ corresponding to the measured experimental parameters.

To model the experiment, we start with the one-dimensional quantum-mechanical equations of motion for SRS, valid because the pump collimation ensures that the Stokes light is nearly diffraction limited [6,7],

$$\frac{\partial}{\partial z}\hat{E}^{(+)}(z,t)=i\kappa_2\hat{Q}(z,t)E_p^*(t)\,, \tag{1}$$

$$\frac{\partial}{\partial t}\hat{Q}(z,t)=-\Gamma\hat{Q}(z,t) -i\kappa_1 E_p(t)\hat{E}^{(+)}(z,t)+\hat{F}(z,t)\,. \tag{2}$$

Here $\hat{E}^{(+)}(z,t)$ is the Stokes field operator, $\hat{Q}(z,t)$ is the creation operator for the molecular vibrations at z, and t is the retarded time. $E_p(t)$ is the pump field which we take to be classical, undepleted, and Gaussian in time. $\hat{F}(z,t)$ is the Langevin operator that is necessary to enforce the proper commutation relations in the presence of collisional damping. We assume that $\hat{F}(z,t)$ has the following correlation functions [7]:

$$\langle\hat{F}^\dagger(z,t)\hat{F}(z',t')\rangle=(2\Gamma/\rho)[1+\bar{n}(z,t)]\delta(z-z')\delta(t-t')\,, \tag{3a}$$

$$\langle\hat{F}(z,t)\hat{F}^\dagger(z',t')\rangle=(2\Gamma/\rho)\bar{n}(z,t)\delta(z-z')\delta(t-t')\,. \tag{3b}$$

The number of molecules per unit length is ρ and $\bar{n}(z,t)$ is the fraction ($\ll1$) of molecules that are vibrationally excited at the time t in a thin slice centered at z.

The leading Stokes field [i.e., that arising from the leading pump pulse $E_{p1}(t)$] is given by [7]

$$\hat{E}_1^{(+)}(z,t)=i\kappa_2 E_{p1}^*(t)\int_{-\infty}^{t}dt'\,e^{-\Gamma(t-t')}\int_0^z dz'\,K_1(z,z';t,t')\hat{F}(z',t')\,, \tag{4}$$

where $K_1(z,z';t,t')$ is a kernel which describes the gain encountered by the propagating Stokes field. The free-field operator $\hat{E}^{(+)}(0,t)$ has been neglected in the solution because the Stokes field is initially in the vacuum state and it will not contribute to detected intensities.

This expression for the field operator shows how the incoherent Langevin fluctuations can initiate a stimulated cascade of scattering events which gives rise to a coherent Stokes field in the large-gain, transient limit. Essentially, because of the high gain, the first few spontaneous scattering events occurring along the direction of the pump beam and near the entrance of the Raman cell will initiate a series of stimulated events that dominate the scattering process. Provided the duration of the pump pulse is short enough that a steady state is not reached ($\Gamma\tau_p < g_0 I_p L$), the phase of the generated field will be determined by those few events which initiated the process. It is in this sense that the resultant Stokes field can be thought of as a macroscopic manifestation of quantum zero-point noise.

The molecular coherence that is present for times between the two pump pulses can be shown to be $\hat{Q}(z,t) = \hat{Q}_C(z,t) + \hat{Q}_U(z,t)$, where

$$\hat{Q}_C(z,t) = e^{-\Gamma(t-t_0)} \int_{-\infty}^{t_0} dt' e^{-\Gamma(t_0-t')} \left[\hat{F}(z,t') + \int_0^z dz' \hat{F}(z',t') \frac{\partial}{\partial z} K_1(z,z';t,t') \right], \tag{5a}$$

$$\hat{Q}_U(z,t) = \int_{t_0}^{t} dt' e^{-\Gamma(t-t')} \hat{F}(z,t') . \tag{5b}$$

The time at which the leading pump pulse ends is denoted by t_0. $\hat{Q}_C(z,t)$ represents the molecular vibrations that are present during the leading pump pulse. The amplified contribution contains K_1 and strongly dominates over the freely evolving zero-point noise $\hat{F}(z,t')$. As a consequence, the phase of this induced vibration will be well defined on any given shot and correlated throughout the medium, although it will vary randomly from shot to shot. In fact, in the limit of high gain the phase of the induced vibrational coherence will be locked to that of the leading Stokes field $\hat{E}_1^{(+)}$. After the leading pump pulse ($t > t_0$) this coherent term decays by collisional dephasing $\exp[-\Gamma(t-t_0)]$, and uncorrelated zero-point noise is introduced in the vibrations via $\hat{Q}_U(z,t)$, the integral of $\hat{F}(z,t')$ from t_0 to t.

Subsequently, the trailing pump pulse initiates a scattering process which gives rise to a trailing Stokes field, given by $\hat{E}_2^{(+)} = \hat{E}_{2C}^{(+)} + \hat{E}_{2U}^{(+)}$, where

$$\hat{E}_{2C}^{(+)}(z,t) = i\kappa_2 E_{p2}^*(t) \int_0^z dz' K_2(z,z';t,t_0) \hat{Q}_C(z',t) , \tag{6a}$$

$$\hat{E}_{2U}^{(+)}(z,t) = i\kappa_2 E_{p2}^*(t) \int_{t_0}^{t} dt' e^{-\Gamma(t-t')} \int_0^z dz' K_2(z,z';t,t') \hat{F}(z',t') . \tag{6b}$$

$\hat{E}_{2C}^{(+)}$ represents the contribution to the trailing Stokes field that is due to the scattering from the molecular coherence $\hat{Q}_C(z,t)$ that is present during the leading pump pulse, while $\hat{E}_{2U}^{(+)}$ is due to scattering from an independent and uncorrelated set of zero-point fluctuations that has built up from the action of the Langevin operator after the leading pump pulse. Hence, these two components will be uncorrelated both in magnitude and in phase. This allows us to model the trailing Stokes field in terms of two independent phasors as shown in Fig. 3. In the limit of high-gain, transient SRS, the field E_{2C} will have a phase that is locked to the phase of the leading Stokes pulse E_1. The relative phase between the leading and trailing Stokes fields is then determined by the vector sum of these two phasors.

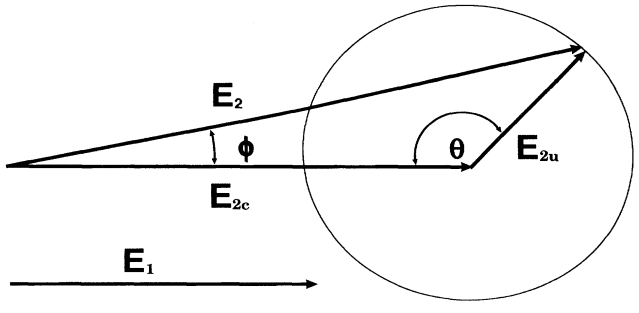

FIG. 3. Phasor representation of the correlation between the complex Stokes fields E_1 and E_2 created by the leading and trailing pump pulses, respectively.

The statistics for the relative phase between E_1 and E_2 can be developed from the statistics of the individual fields E_{2C} and E_{2U}. Because E_{2C} and E_{2U} are assumed uncorrelated, the phase θ in Fig. 3 will be uniformly and randomly distributed on the interval $(-\pi,\pi)$. Furthermore, it is known that the magnitude of the Stokes field in the transient regime is well described by a single temporal mode function with a fluctuating amplitude that obeys Gaussian statistics [7]. Although the temporal modes for E_{2C} and E_{2U} are slightly different, owing to different time dependences in the propagators K_1 and K_2 (E_{2C} starts to rise earlier in time than E_{2U}), the shapes of the peaks are similar. Since the calculation of the phase variance is dominated by the peak values, we ignore the different temporal dependences. The statistics of E_{2C} and E_{2U} depend solely on their variances which can be calculated from Eq. (6).

The theoretical result for the variance of the relative phase between the leading and trailing Stokes pulses is shown in Fig. 2 for two different values of the gain. The curve labeled with a gain of 75 is that expected using measured experimental parameters. The curve corre-

sponding to a gain of 90 fits the data somewhat better. This may reflect the error in the measurement of the Raman gain due to fluctuations in the pump laser.

It is important to develop a quantitative measure of how near the surviving coherent excitation is to the quantum-noise limit. In the case of light injection into an interferometer with internal gain, the measure is naturally given in terms of numbers of photons per mode [1-3]. This motivates in our case the introduction of a description of the surviving molecular coherence in terms of delocalized vibrational quanta. In a crystal these would be referred to as optical phonons [9], and we will use this terminology here with the understanding that many typical properties of phonons are absent in a gaseous medium.

We start by defining an optical-phonon creation operator, again in the one-dimensional approximation [7], as

$$\hat{a}_m^\dagger = (N_0^{1/2}/L)\int_0^L dz \exp(-i2\pi mz/L)\hat{Q}(z,\tau), \quad (7)$$

where N_0 is the number of H_2 molecules in the interaction (and quantization) volume, of length L, and $m=0, 1, \ldots, N_0-1$. The number of phonon modes equals the number of molecules, while the wave number of mode m is given by $(\omega_p-\omega_s)/c+2\pi m/L$. These operators obey the usual Bose commutation relations.

The average total number of phonons excited during the leading pump pulse, ending at time t_0, is

$$N_{\text{phonon}} = \sum_{m=0}^{N_0-1}\langle \hat{a}_m^\dagger \hat{a}_m\rangle = \int_0^L dz\, \rho\langle \hat{Q}_C(z,t_0)\hat{Q}_C^\dagger(z,t_0)\rangle, \quad (8)$$

where $\rho=N_0/L$. Neglecting any collisional dephasing occurring during the pump pulse, these phonons will all be correlated at t_0 due to the coherent nature of the transient scattering process. The initial excitation of phonon modes will be determined by the spatial distribution of the excited molecules $\hat{Q}_C(z,t_0)$, which has the form $\exp(g_0 I_p \Gamma \tau_p z)^{1/2}$ in the transient, high-gain limit. The fraction in the lowest-order mode depends on the gain-length product. After dephasing collisions have occurred for a time period τ the total number of phonons is constant, but the average number N_C that remain correlated to the initial set decays as $N_C = N_{\text{phonon}}\exp(-2\Gamma\tau)$, which follows from the time evolution in Eq. (5a). Eventually dephasing collisions will lead to all modes being equally excited and mutually incoherent. Since the number of modes $N_0 \approx 10^{20}$) is far greater than the initial number of phonons ($N_{\text{phonon}} \approx 10^{10}$) this final excitation is well below the zero-point noise, whose strength corresponds to one phonon per mode.

To estimate how close the present experiment is to this quantum-noise limit, we use energy conservation to equate the average initial number of phonons to the average total number of Stokes photons created by the leading pump pulse. This number was measured to be between 3×10^9 and 1×10^{10}, the accuracy of which was limited by the 10% fluctuations of the pump laser power. The longest delay τ for which we detected a normalized phase variance less than 0.85, the bound set by the interferometer jitter, is 8.8 collisional dephasing times. This corresponds to between 70 and 230 surviving correlated phonons. We estimate theoretically that the $m=0$ mode contains about 22% of the phonons, or between 15 and 50 phonons at this delay. Other modes have fewer phonons.

In conclusion, we have measured the variance of the relative phase of two interfered Stokes fields generated from the same molecules at different times. The phases are found to be well correlated for time delays of approximately nine collisional dephasing times. This interferometric method allows one to study the response of the SRS process to initial molecular coherence near the limit set by the zero-point vibrational motion. With an improved technique, it should be possible to detect of the order of one phonon per mode. This new technique could be applied to the detection of small numbers of phonons in condensed media that are Raman or Brillouin active.

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