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# Proposal for the Measuring Molecular Velocity Vector with Single-Pulse Coherent Raman Spectroscopy*

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**Abstract.** Methods for simultaneous measurements of more than one flow velocity component using coherent Raman spectroscopy are proposed. It is demonstrated that using a kilowatt broad-band probe pulse (3-30 GHz) along with a megawatt narrow-band pump pulse (~100 MHz), coherent Raman signal resulting from a single laser pulse is sufficient to produce a high-resolution Raman spectrum for a velocity measurement.

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Now that the proposed method of using coherent Raman spectroscopy for molecular flow measurements [1] has been experimentally demonstrated with stimulated Raman gain [2] (SRGS), coherent anti-Stokes Raman [3] (CARS) and inverse Raman [4] (IRS) spectroscopies, it is time to consider exact requirements in most practical applications. Although the signal-to-noise in the recent IRS experiment [4] is quite good, the demonstration experiments up to this point have dealt with the measurement of one velocity component (along the direction of the overlapping laser beams) averaged over the entire interaction region for a time of several minutes. The relatively long measurements time (5 or 10 min) is the result of the need to frequency scan one single-frequency laser beam to obtain the desired Raman spectrum. In a typical application of flow-velocity measurements, several improvements from the present achievements are desired or required: 1) higher spatial resolution and simultaneous multiple-point measurements would be useful for flow analysis and evaluation, 2) more than one velocity component is needed to determine the flow velocity vector, and 3) a much shorter measurement time would be preferred if many velocity measurements are to be made or if the behavior of turbulence is to be studied (in a wind tunnel, for example). In this article a method for measuring two velocity components and techniques for obtaining the desired coherent Raman spectrum on a single-pulse basis are proposed. The available signal and signal-to-noise (S/N) will be considered and the possibility of simultaneous multiple point flow velocity measurements discussed.

There are four types of coherent Raman spectroscopy [5]. Besides the SRGS, CARS, and IRS mentioned, there is the coherent Stokes Raman spectroscopy (CSRS), in which the probe beam is at the anti-Stokes frequency and the emitted signal beam is at the Stokes frequency. For simplicity, we consider IRS and CSRS, with the probe beams at the anti-Stokes frequency $\omega_a$, and wave vector $\mathbf{k}_a$. The results obtained can be applied to SRGS and CARS by interchanging the Stokes frequency and wave vector $\omega_s, \mathbf{k}_s$ with $\omega_a, \mathbf{k}_a$. The Raman resonance occurs for both IRS and CSRS for a moving molecule with velocity $\mathbf{v}$ when

$$\omega_a = (\omega_l + \Delta) + (\mathbf{k}_a - \mathbf{k}_l) \cdot \mathbf{v}, \tag{1}$$

where $\Delta$ is the molecular vibration frequency and $\omega_l$ and $\mathbf{k}_l$ are the frequency and wave vector of the pump beam. Equation (1) can obviously be used to determine the velocity component pointing in the direction $\hat{\varepsilon} \parallel (\mathbf{k}_a - \mathbf{k}_l)$ using IRS. For CSRS, the resonance in the scattering cross section occurs at the same frequency difference $\omega_a - \omega_s$, and the velocity component along $\hat{\varepsilon}$ is given by (1) also. Note that, in addition, the phase matching condition, $\mathbf{k}_a - \mathbf{k}_l = \mathbf{k}_l - \mathbf{k}_s$, must be satisfied for CSRS.

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## Measuring Two-Velocity Components

When two fixed frequency pump beams with $\omega_l, \mathbf{k}_l$ and $\omega_l, \mathbf{k}'_l$ and one tunable probe beam with $\omega_a, \mathbf{k}_a$ are used, resonance in Raman cross-section will occur at two anti-Stokes frequencies. They are given by (1) and

$$\omega'_a = (\omega_l + \Delta) + (\mathbf{k}_a - \mathbf{k}'_l) \cdot \mathbf{v}. \tag{2}$$

Equation (2) can then be used to determine, independently, the velocity component pointing along $\hat{\varepsilon}' \parallel (\mathbf{k}_a - \mathbf{k}'_l)$. Both resonances can be probed by either IRS or CSRS. If CSRS signals are sought for in this arrangement, the three-dimensional phase matching condition [6, 7] must be met and the Stokes signal may be detected along $\mathbf{k}_s = \mathbf{k}_l + \mathbf{k}'_l - \mathbf{k}_a$. As the probe frequency is tuned, resonances at $\omega_a$ and $\omega'_a$ are measured from which the velocity components along $\hat{\varepsilon}$ and $\hat{\varepsilon}'$ are determined. In experiments in which the pump frequency is varied, $\omega'_a$ and $\omega_l$ in (2) should be replaced by $\omega_a$ and $\omega'_l$. For stationary molecules, $\mathbf{v} = 0$, the two resonance peaks merge into one. Although two velocity components can be measured this way, there is a theoretical ambiguity in assigning resonance peaks to the correct velocity components. In practice, one may align (more or less) the main flow along the $\hat{\varepsilon}$ direction, then the larger Doppler shift corresponds to this velocity component. How well the two peaks can be resolved, of course, depends on the Raman linewidth and the signal to noise. If one more beam (probe or pump) is properly added, with a total of four beams at two different frequencies, three velocity components can be measured simultaneously. This is not an over-complicated setup as three-beam CARS has been used routinely for Raman thermometry measurements [8, 9].

## Single-Pulse Measurements

In order to reduce the measurement time, pulsed coherent Raman experiments using narrow-band pump beam (or beams) and a broad-band probe beam are proposed. The idea here is similar to the existing broad-band CARS thermometry experiments [8, 9], except that the probe bandwidth should now be about 5–10 GHz and the pump bandwidth be narrower than 100 MHz [4, 10]. Since the signal obtained from a single pulse under proper conditions should be strong enough for recording a Raman spectrum, high-resolution and braod-band single-pulse CSRS (or CARS) experiments are proposed for flow-velocity measurements. Unlike IRS, CSRS signal contains no dc background and the emitted Stokes Raman spectrum may be recorded by an optical multi-channel analyzer (OMA). If the proposed single-pulse Raman experiment for measuring flow velocities is to be successful, one must make sure that (a) the CSRS signal is strong enough to be recorded by the diode arrays in an OMA, (b) a probe laser of 5–10 GHz bandwidth with reasonably stable, uniform and reproducible spectrum can be constructed, and (c) a spectrum analyzer with sufficient resolution to resolve the CSRS signal (Q-branch vibration Raman lines of $N_2$ for example), is available. Although some of these items do not yet exist, they can be constructed with present technology.

Assuming that the necessary probe laser and spectrum analyzer exist, the CSRS signal resulting from Q-branch $N_2$ lines can be shown to be strong enough for recording. At visible wavelengths, the stimulated Raman gain (or loss) of a 15% populated line ($J = 8$ at room temperature, for example) in the Q-branch $N_2$ Raman spectrum at atmospheric pressure with 3 GHz linewidth is calculated to be $G = 10^{-7}$ per 1 W of the pump power [1]. This assumes that the pump and probe beams overlap and have Gaussian profiles. Under the phase matched conditions, the CSRS signal power resulting from monochromatic pump and probe beams can be estimated approximately from

$$P_s \simeq G^2 P_a. \tag{3}$$

If the width of the pump laser is 100 MHz with 1 MW power, and the probe power is $P_a = 1$ W, then the CSRS signal in the 100 MHz band is calculated to be approximately $P_s \simeq 10^{-2}$ W. With a 10 ns pulse width, the signal per pulse is about $3 \times 10^8$ photons in the 100 MHz band. If a 5 GHz probe laser is used, the total power of the probe pulse required is 50 W. To analyze this CSRS signal, a Fabry-Perot spectrometer with free spectral range of 5 GHz and finess of 50 which yields a spectral resolution of 100 MHz may be used. Assuming three Fabry-Perot interference orders are recorded and a factor of 100 for quantum efficiency and optical losses, the signal strength of a detected order per 100 MHz is still $10^6$ photons per laser pulse. If all these photons are deposited in one OMA channel, the shot-noise limited S/N would be $10^3$! If these photons are divided into 10 channels, the S/N in one channel is still 300. Other typical situations and the associated parameters are given in Table 1. Obviously the signal strength is adequate for all these cases. Since the coherent Raman signal per 100 MHz band is independent of gas pressure if pressure broadening dominates, and the Raman linewidth is much narrower [4] than 3 GHz at pressures lower than 1 atm, the calculated signal strength may be considered typical. Because the nitrogen molecule is a relatively weak Raman scatterer, more signal could be expected, if other Raman scatterers are of interest.

Table 1. Calculated single-pulse CSRS experimental parameters [1 MW pump beam (100 MHz linewidth and 10 ns pulse width), 10 ns probe pulse, a finess of 50 for the spectral analyzer and complete overlap between pump and probe beams are assumed]

| Probe power [W] | Laser spectrum width [GHz] | Spectrum analyzer free spectral range [GHz] | Frequency resolution [MHz] | Detected signal per 100 MHz channel [photons] |
|---|---|---|---|---|
| 30 | 3 | 3 | 60 | $10^6$ |
| 50 | 5 | 5 | 100 | $10^6$ |
| 100 | 10 | 10 | 200 | $10^6$ |
| 300 | 30 | 30 | 600 | $10^6$ |

## Suggested Implementation

Although the needed probe-pulse laser and the high-resolution spectrum analyzer are not available commercially, one is certain of their realization within current technology. A Fabry-Perot interferometer [11] with high enough finess is an obvious candidate for the required high-resolution spectrum analyzer. Alternatively, Fizeau interferometry [12] can also be used to record the coherent Raman spectra. In general, any schemes with high-enough resolution ($\sim 100$ MHz) for measuring the wavelengths of a pulsed laser can be used with an intensified OMA to perform the required spectral analysis. Unlike the pulsed wavemeter [11], the analyzer (interferometer) for this proposed application, needs to make a relative frequency measurement only. The optical portion of the device should be much simpler.

Although a light source with no sharp frequency structure (laser modes) within the frequency band of interest, as given in Table 1, has not been made, one may start with the spontaneous emission from a dye cell and then properly filter its spectral content with a grazing-incidence grating in connection with an etalon or atomic filter [13] when necessary. After the spontaneous emission with proper linewidth is obtained, amplifier stages may be used to obtain the desired power. The spectral output of the dye amplifier then be investigated on a pulse-to-pulse basis to ensure the approximate spectral stability and uniformity of the probe pulse. However, the absolute frequency of Raman resonances is determined by the frequency stability of the narrow-band pump, a slight variation in the smooth probe spectrum profile will not affect the result of the velocity measurements. Since the required probe power is lower than 1 kW per pulse (Table 1), a pulsed YAG laser should have enough power to pump this broad-band probe source as well as the narrow-band dye amplifier for the pump. If the probe wavelength is chosen to be around 5145 Å, as used in the IRS work [4], one needs to use the third harmonic of the YAG laser to pump the C 500 dye to generate the desired probe beam. Alternatively, it may be better to use the second harmonic of the YAG laser at 5320 Å to pump both lasers at, for example, 5560 and 6388 Å. The dyes to be used would be Rhodamine 6G and Rhodamine 640, respectively. One may also perform single-pulse CARS experiments, if the 6388 Å is picked for the probe. In this case, the single-mode cw dye laser beam (using Rhodamine 110 dye) at 5560 Å would be amplified to produce the narrow-band pump pulses. Although a broad-band pump and narrow-band probe pulse can also work, it is not a good practice to use a broad-band pump because too much power would be wasted.

The necessary electronics for the interface of OMA and minicomputer required for the proposed single-pulse coherent Raman experiment has been developed and implemented in a similar experiment for CARS thermometry [9]. Although much higher spectral resolution is required for the Raman velocimetry proposed, the requirements on the sensitivity of the intensified diode array, and on the data acquisition electronics are the same. In fact, unlike the rotational Raman thermometry work [9], there is no need to monitor the probe spectrum profile on pulse-to-pulse basis for velocity measurements, thus simplifying the required electronics.

The optical arrangement for spatially resolved multiple-point coherent Raman experiments is straightforward and such measurements on a $CH_4$ concentration distribution have been made before [14]. The question at hand is whether we have enough Raman signal to be divided into spatially resolved points. Consider, for example, the recent frequency scanned IRS experiment [4]. An approximate $S/N \simeq 40$ was obtained with 1 MW pump and 300 mW probe. If one uses a cylindrical focussing lens and attempts to have 10 spatially resolved channels, one has the same S/N with 10 times the pump and probe powers or with 100 times the probe power and the same pump power. This should certainly be possible. For the single-pulse CSRS experiments given in Table 1, ten spatially resolved points with the same total pump and probe power would yield a reduction of signal photons by a factor of $10^3$. This still leaves one with $10^3$ photons in every 100 MHz channels of every spatially resolved point. The shot-noise limited signal-to-noise in each channel is then about 30, not an unworkable number. Although the SRGS and IRS experiments with overlapping pump and probe beams have yielded measured signals to about a factor of 5 from the theoretical value [2, 4], CARS experiments apparently have a much larger discrepancy [14]. If such a discrepancy (2 to 3 orders of magnitude) which is not fundamental, can not be eliminated or if the probe and pump beams are crossed for higher spatial resolution, then certain amount of pulse averaging may be necessary. In that event, laser systems with higher repetition rate should be considered [1].

## Conclusion

In summary, it is theoretically demonstrated that velocity measurements using coherent Raman spectroscopy can be implemented to simultaneously handle more than one velocity component in multiple spatially resolved points with current technology. Such simultaneous velocity measurements can be carried out on single-pulse basis with a measurement time of the laser pulse duration (less than 10 ns). When higher spectral resolution, longer-pulse-duration laser systems [15] and multi-channel transient digitizing techniques become highly developed, spatial distribution of velocity vectors can be measured as a function of time with microsecond resolution [16]. Such an instrument would then be ideally suited for high flow speed and turbulence measurements. Although the proposed techniques of coherent Raman spectroscopies are perhaps technically more involved than other velocimetry methods [17], this is the only technique that requires absolutely no seeding and that offers promises for measuring high-speed flows unmatched by any other known method.

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