**V. V. Smirnov,** *Coherent Raman Spectroscopy of Gases.* Progress in the development of tunable laser sources of light has led, in the last few years, to the development of coherent Raman spectroscopy. This method is based on the wavelength shift in media with cubic nonlinearity of susceptibility $\chi^{(3)}$, the possibility of which was experimentally demonstrated for the first time by Maker and Terhune.[1] In this process, the application of spatially coherent fields due to two powerful lasers with frequencies $\omega_1$ and $\omega_2 = \omega_1 - \Omega$, where $\Omega$ is the frequency of the Raman-active transition under investigation, leads to the biharmonic excitation of oscillations in the medium. The biharmonic "pump" produces high excited-state populations and a phasing of the oscillations of the medium throughout the volume in which the interaction takes place. The intensity of the pumping radiation scattered by a medium of this kind is much greater than the intensity resulting from spontaneous scattering, but at the same time there is no competition between the lines because the threshold for stimulated Raman scattering is not reached. By varying the frequency of one of the lasers, and thus varying the difference $\omega_1 - \omega_2$, it is possible to excite and investigate a series of Raman-active oscillations. In practice, one records the signal due to the coherent anti-Stokes scattering of light. Another important feature is that the scattered radiation is localized within the diffraction angle, i.e., it is laser-like, in contrast to spontaneous Raman scattering, where the scattering indicatrix is isotropic. To increase the scattered intensity, one must ensure that the momentum conservation law

$$2\omega_1 - \omega_2 = \omega_a, \qquad 2\mathbf{k}_1 - \mathbf{k}_2 = \mathbf{k}_a,$$

is satisfied in addition to energy conservation. In the case of collinear propagation, the effective interaction occurs over the coherence length, i.e., the length of phase mismatch determined by the dispersion of the medium ($\Delta k L_{coh} \approx \pi$). The characteristic coherent lengths in liquids and solids are of the order of a few millimeters or a fraction of a millimeter, whereas, in gases at normal density, they amount to hundreds of millimeters. Simple calculations show that even the use of lasers with relatively low output power (~ 1 kW) can produce an increase in the scattered intensity by a factor of at least $10^9$.

The above features of coherent anti-Stokes scattering of light make this method ideal for the detection of Raman-active spectra in highly luminescent media such as, for example, biological objects, in research on plasmas and discharges, and in photochemistry. The

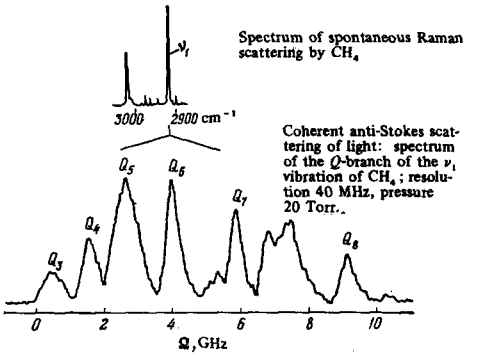


FIG. 1.

possibilities of this method in Raman spectroscopy have been demonstrated in a number of investigations involving solids,[2,3] liquids,[4,5] and gases.[6–10]

This paper is mainly concerned with results demonstrating the possibilities of coherent anti-Stokes scattering of light in the spectroscopy of vibrational-rotational transitions in gases. For example, the rotational and vibrational temperatures have been determined[6–8] for gas discharges and flames, and the topography of individual fragments in gas flames has been established with a spatial resolution of about 20 $\mu$m.

One of the fundamental problems in Raman spectroscopy is the question of spectral resolution. Resolution of no better than 0.1 cm$^{-1}$ can be achieved in the detection of spontaneous Raman scattering in specialized laboratory systems, using exposures of the order of some tens of hours. On the other hand, in coherent anti-Stokes scattering of light, the resolution is determined by the laser linewidth. The use of single-frequency, continuously operating, lasers has resulted[9,10] in the record resolution of 40 MHz (0.001 cm$^{-1}$), and the spectra of the $Q$-branches of a number of the molecular vibrations have been completely resolved (see Fig. 1).


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