Generation of coherent IR light on a dipole-forbidden molecular transition with biharmonic pumping in a static electric field

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Coherent IR emission from the homonuclear molecule H_2 has been induced by laser light and a static electric field. This effect might be utilized to develop a nonlinear-optics method for remote measurements of electric fields with spatial and temporal resolution.

Vibrations of homonuclear molecules are inactive in the IR part of the spectrum in the absence of external agents. In the present study we have achieved coherent IR emission from the H_2 molecule. To achieve this emission, we modified the method of coherent active Raman spectroscopy (CARS). The standard layout[1] for coherent scattering is shown in Fig. 1a. The interaction of three waves (with frequencies ω_1, ω_2 and ω_3) with the medium results in the generation of coherent light at the frequency $\omega' = \omega_1 - \omega_2 + \omega_3$. A static electric field can be used in place of one of the interacting waves. Several studies (e.g., Refs. 2 and 3) have been carried out on the frequency conversion of laser beams by means of a cubic nonlinearity of a medium in a static electric field. For the most part, these studies have dealt with the generation of the sum frequency of coherent light.

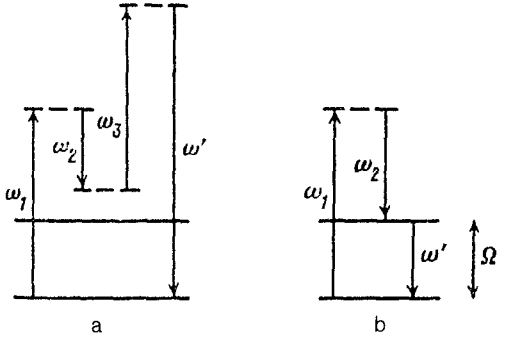

FIG. 1. Possible schemes of processes involving the cubic nonlinearity of a medium. a—Generation of light at the frequency $\omega' = \omega_1 - \omega_2 + \omega_3$; b—generation of light at the difference frequency $\omega' = \omega_1 - \omega_2$ in a static electric field. Here Ω is the frequency of Raman-active molecular vibrations.

In the present experiments we have implemented a different scheme (Fig. 1b) for the nonlinear interaction of two laser beams and a static electric field with a medium. In the case at hand, this medium consists of hydrogen molecules. In these experiments, in contrast with the approach outlined above, we detect the generation of coherent light at the difference frequency $\omega' = \omega_1 - \omega_2$, rather than the sum frequency, in a static electric field. According to Fig. 1b, the intensity of the signal at the frequency ω' in a static electric field **E** can be written

$$I(\omega' = \omega_1 - \omega_2) \propto |X^{(3)}(\omega_1 - \omega_2)|^2 \mathbf{E}^2 I_1 I_2, \qquad (1)$$

where $X^{(3)}$ is the cubic susceptibility of the medium, and $I_{1,2}$ are the intensities of the laser beams. Another distinguishing feature of these experiments is that the difference $\omega_1 - \omega_2$ is tuned to resonance with the frequency (Ω) of a Raman-active transition. This approach results in a resonant growth of the cubic susceptibility of the medium, $X^{(3)}$.

The experiments on the generation of IR light by H_2 molecules in a static electric field E were carried out in a pressure chamber (the pressure was varied up to 10 atm). A capacitor with an air-filled gap 3.5 mm wide was in this chamber. The voltage applied to the capacitor plates made it possible to vary the electric field over the range $E = 0$–45 kV/cm. The layout of the CARS spectrometer used here is described in Ref. 4. The wave with frequency ω_1 ($\lambda_1 = 532$ nm) was the second harmonic from a pulsed ($\tau_i = 10$ ns) Nd:YAG laser with a pulse repetition frequency of 20 Hz. The energy in the pulse was $J_1 = 12$ mJ. A tunable dye laser (the dye was pyridine-1) generated the light with ω_2 ($\lambda_2 \sim 683$ nm), which satisfied the Raman resonance condition $\omega_1 - \omega_2 = \Omega$ for the $v = 0$, $v = 1$ transition of the $H_2(X\,^1\Sigma_g^+)$ molecule. The energy in this pulse was $J_2 = 1$ mJ. The field E and the fields of the linearly polarized light with ω_1 and ω_2 were in the same direction. We used a collinear arrangment to bring the ω_1

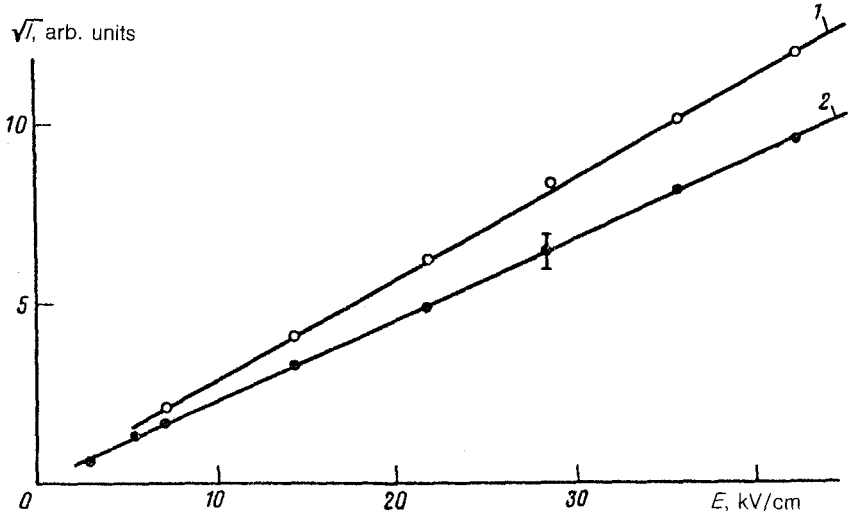

FIG. 2. Experimental results on the intensity of the IR emission signal as a function of the strength of the static electric field E for various pressures of molecular hydrogen. *1*—10 atm; *2*—8 atm.

and ω_2 beams into coincidence. These beams were focused in the region between the capacitor plates. A signal at the frequency $\omega' = \omega_1 - \omega_2$, with a wavelength $\lambda \sim 2.4$ μm, was tapped from the laser beam by an LiF prism and was detected by an IR detector based on a liquid-nitrogen-cooled InSb photodiode. A Ge window at the entrance to this detector blocked scattered visible light. The output signal from the detector was fed to a boxcar integrator and then to a chart recorder.

The intensity of the light with $\lambda \sim 2.4$ μm had a quadratic dependence on the strength of the static electric field E. Figure 2 shows some representative results on the behavior of $\sqrt{I}$ as a function of E for the line $Q(1)[v'' = 0, J'' = 1 \rightarrow v' = 1, J' = 1]$ for various hydrogen pressures. We see that the plots are linear, i.e., they correspond to relation (1). At an H_2 pressure $p = 10$ atm, the minimum field E at which the 2.4-μm signal was detected under these experimental conditions was ~ 3 kV/cm. Our estimates indicate that appropriate refinements of the apparatus could raise the sensitivity substantially.

We wish to stress that the light with $\lambda \sim 2.4$ μm had a narrow directional pattern, and its intensity was a quadratic function of the pressure (the pressure was varied over the range $p = 6$–10 atm).

These experiments have thus resulted in the generation of coherent IR light on a dipole-forbidden molecular transition during biharmonic pumping in a static electric field. Since this transition belongs to the Q branch, it is not observed in the absorption spectrum (or in the emission spectrum) in the absence of a static electric field, in either homonuclear or heteronuclear diatomic molecules. The reason lies in selection rules on the rotational quantum numbers. In addition to its nonlinear-optics applications, this scheme (Fig. 1b) might be turned into a method for spatially and temporally resolved remote measurements of electric fields in gases and plasmas.[1)] Here are the characteristic features of this method:

1. The laser apparatus required for the implementation of the scheme in Fig. 1b is widely available. In particular, it is available in any type of CARS apparatus.

2. In the scheme in Fig. 1b, the generation of the "ordinary" degenerate ($\omega_3 = \omega_1$) CARS signal at the frequency $\omega_a = 2\omega_1 - \omega_2$ is observed simultaneously. This signal can be used to monitor the particle density in the measurement zone. This capability is particularly important in work with spatially inhomogeneous objects. It also substantially simplifies the process of adjusting the overall system.

3. The scheme which we are discussing here can be used for local measurements in oscillating as well as static electric fields. If the frequency of the oscillating field, $\omega > \tau_i^{-1}$, is in the microwave range, one may achieve generation of IR light at two closely spaced frequencies $\omega' = \omega_1 - \omega_2 \pm \omega$ instead of generation of IR light at the frequency $\omega' = \omega_1 - \omega_2$. By analogy with (1), the intensity of the light, $I(\omega')$, would be proportional to the square of the amplitude of the oscillating electric field.

The working molecules need not be H_2; they could also be such molecules as N_2, CO_2, H_2O, and CO.

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[1)]Buldakov *et al.*[5] have demonstrated theoretically that it is possible to measure a static electric field in a molecular-gas medium by coherent active spectroscopy. However, they stipulated that the frequency of one of the laser beams be the same as the frequency (Ω) of the Raman-active vibrations of the scattering molecules.[5] That stipulation complicates efforts to implement their suggestion in practice.

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