Power spectrum of coherent Rayleigh-Brillouin scattering in carbon dioxide

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We show in this note that, in the coherent Rayleigh-Brillouin scattering (CRBS) experiment [X. Pan, M. N. Shneider, and R. B. Miles, Phys. Rev. A **69**, 033814 (2004)], the vibrational modes of the CO_2 molecules are frozen. When the gas dynamic parameters are chosen accordingly, the model predicts a line shape that matches with the experimental data. Fitting the theoretical curve to the CRBS data represents a method to measure the speed of high-frequency sound, bulk viscosity, and the rotational relaxation time of molecular gases.

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In this note, we report a further study on the CO_2 coherent Rayleigh-Brillouin scattering (CRBS) power spectrum. We realize that, in the experimental conditions, the CO_2 vibrational modes are frozen. When the gas parameters are appropriately chosen for this physical condition, our model in [1] correctly predicts the speed of sound and matches the CRBS power spectrum of CO_2.

As described in [1], in a CRBS experiment, two counter-propagating pump laser beams, polarized in the same direction, are focused and crossed at their foci. They form an interference pattern and generate a wavelike density perturbation in the gas. A probe beam is then coherently scattered from the perturbation and forms the CRBS signal. The observed power spectrum of CRBS in CO_2 is shown in Fig. 1

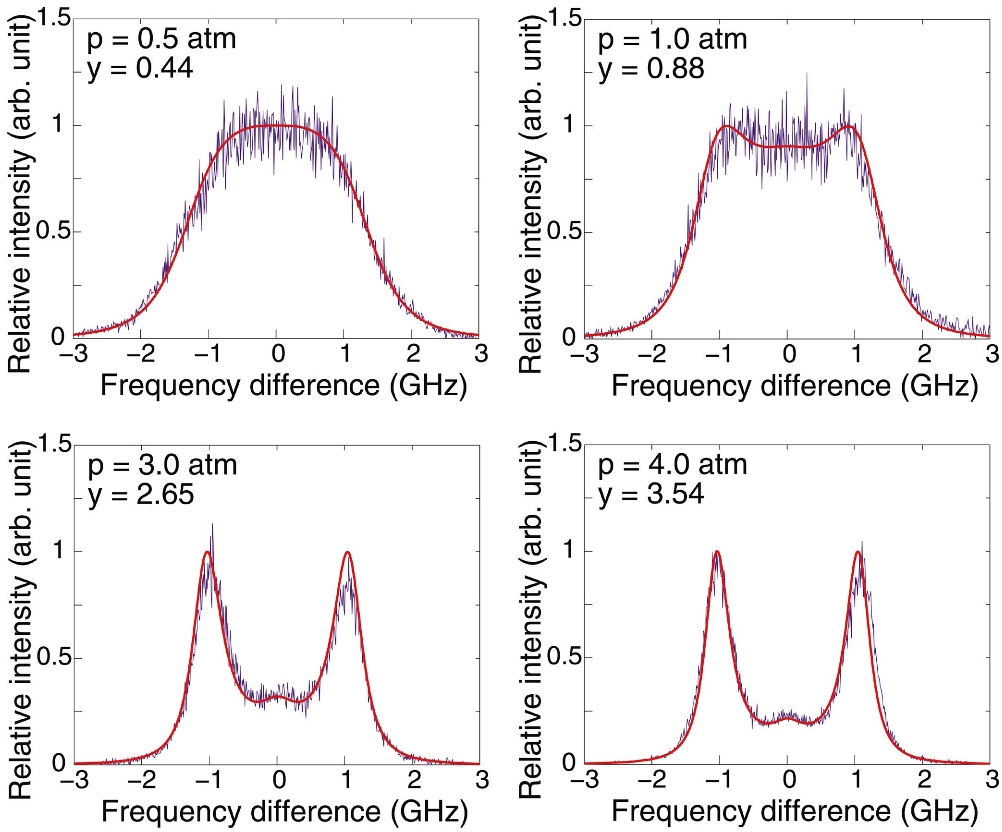

FIG. 1. (Color) Coherent Rayleigh-Brillouin scattering in carbon dioxide at $T = 292$ K. The thin curves are the experimental data; the thick curves are the theoretical line shape. The theoretical curves are calculated using $\eta_b = 0.25\eta$.

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with the same set of data as appeared in [1]. (The vertical axis is rescaled to match the present model curve at the center of the line shape.) These data were taken at a y parameter of approximately the ratio between the scattering wavelength and the mean free path, varying from 0.44 to 3.54. At high pressures, the Brillouin peaks are distinctive; their positions are related to the speed of sound. At temperature $T=292$ K, the measured speed of sound in CO_2 is $v_s=280\pm5$ m/s.

The measured speed of sound agrees almost exactly with the high-frequency asymptotic value given in [2]. Reference [2] shows that the speed of sound increases with the frequency and tends to an asymptotic value of 282 m/s at 10^6 Hz due to the freezing of the vibrational modes. In our CRBS experiments, the frequency is on the order of 1 GHz; therefore, the vibrational modes are frozen. Under this physical condition, we should choose the heat capacity ratio $\gamma=c_p/c_v=1.4$ and the internal heat capacity $c_{\mathrm{int}}=1.0$.

Following the same reasoning, the bulk viscosity is given by $\eta_b=p\tau_r 6r/(3+r)^3$, where $r=2$ is the number of rotational degrees of freedom and τ_r is the rotational relaxation time [3]. The best agreement between the experimental CRBS data and the theory is obtained when we choose $\eta_b\sim0.25\eta$, where η is the shear viscosity. This yields the rotational relaxation time $\tau_r\sim9.3\times10^{-10}$ s, about three times the collisional time. (This result coincides with the transport coefficient measurements in thin capillaries in CO_2 at 300 K [4].) In [1], we used $\eta_b\sim1000\eta$, which is only for low-frequency conditions where vibrational modes couple with translational modes.

In Fig. 1, we plot the theoretical curve together with the experimental data, using $\gamma=1.4$, $c_{\mathrm{int}}=1.0$, shear viscosity $\eta=14.6\times10^{-6}$ Pa s, bulk viscosity $\eta_b=0.25\eta$, and heat conductivity $\lambda=(1/4)(9\gamma-5)\eta c_v$ (Euken relation [3]), where, with vibrational modes frozen, $c_v=(1+3/2)R$, and R is the gas constant. We conclude that the kinetic model developed in [1] can well explain the observed CRBS power spectrum of CO_2. However, in [1], the choices of CO_2's gas-dynamic parameters (γ, c_{int}, and λ) corresponded to the case that the vibrational modes are not frozen; consequently, the model curve did not match the experimental data.

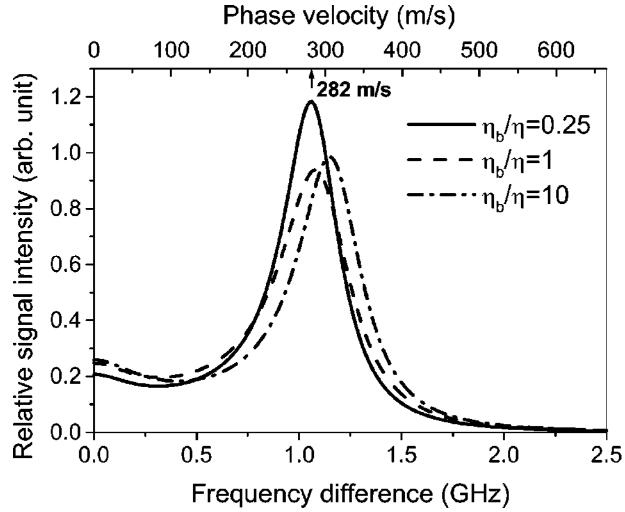

FIG. 2. Computed coherent Rayleigh-Brillouin scattering line shape in CO_2 with different bulk to shear viscosity ratios at $y=3.54$.

In conclusion, we find that in the gigahertz frequency perturbative waves generated in the CRBS experiment, the vibrational modes of CO_2 molecules are virtually frozen. As a result the effective heat capacity ratio $\gamma=1.4$ and the bulk viscosity is about 1/4 of the shear viscosity. This explains the observed CRBS power spectrum and speed of sound. The theoretical model described in [1] is therefore valid for CO_2. We also note that the CRBS data can be used to measure the gas's bulk viscosity, a useful parameter in many cases. In Fig. 2, the model line shapes with different values of η_b are compared for $y=3.54$. We see that the model line shape is sensitive to the value of bulk viscosity. It would be interesting to perform CRBS experiments in high-temperature molecular gases where the vibrational modes couple with the translational and rotational modes.

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