Observation of vibrational motion of adsorbed xenon atoms

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Raman scattering of an argon laser beam by xenon atoms adsorbed on a glass surface is reported.

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In physical adsorption, the atoms and molecules on the surface execute vibrations in an adsorption potential. Observation of these vibrations is important for studying the adsorption interaction. Ordinarily, the relative concentration of adsorbed particles is low, making them difficult to observe, but their number can be increased dramatically by using fine-pore adsorption structures with a greatly extended surface area. If these structures are transparent to a laser beam, optical methods can be used for measurements throughout the adsorbent volume.[1] In order to study the vibrational motion directly in the adsorption potential, we would like to eliminate the effects of internal vibrations of the adsorbed particle, which seriously complicate the observed spectra.[2] For this reason there is particular interest in observing the Raman scattering spectra of adsorbed atoms of inert gases. Even at high concentrations of such atoms in the pore volume, the only atoms that participate in the Raman scattering are those which interact directly with the pore surface.

In the present experiments we have observed Raman scattering of the beam from an argon laser by xenon atoms adsorbed in a "monodispersed" fine-pore glass with an average pore diameter of 80 Å (Ref. 3). We used a DFS-24 Raman spectrophotometer and an LG-68 argon laser with an output power of up to 20 mW in the lines at

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488 and 514 nm. The scattering was observed from the side.

A porous sample with dimensions of 4 X 4 X 10 mm was placed in a vacuum cell, which was then pumped with an NORD-100 vacuum pump for 2 weeks while the sample was baked at 120° C. The vacuum in the cell was (2–4) X 10^{-7} Torr before the cell was filled with xenon. The sample was filled with spectrally pure xenon at a pressure of 600 Torr. After the system had been left for the long time (12 h) required to saturate the sample with xenon, we observed some intense Raman scattering bands which were not exhibited by the pure sample. When the cell was again pumped down for a long time, these bands disappeared, and readmission of xenon completely restored them. Their intensity depends linearly on the laser beam over the range 10–200 mW, amounting to 10^{-6} of the scattering intensity at the laser frequency, measured in the same direction and into the same solid angle. The scattering spectra were identical at the wavelengths 514.5, 488.0, and 496.6 nm.

Figure 1 shows some measured Raman scattering spectra, in which we can see Stokes and anti-Stokes components. Interestingly, the anti-Stokes component is 10–11 times more intense than the Stokes component (spectrogram a in Fig. 1). We do not believe that there is any possibility of a superimposed parasitic line of other origin here. When the same part of the sample was exposed to the laser beam for a long time, the intensity of the anti-Stokes component fell off, and it could fall below the intensity of the Stokes component. Spectrograms a, b, and c were measured after illumination for 10, 20, and 30 h, respectively. When the laser beam was shifted to a different part of the sample, the intensity ratio of the anti-Stokes and Stokes com-

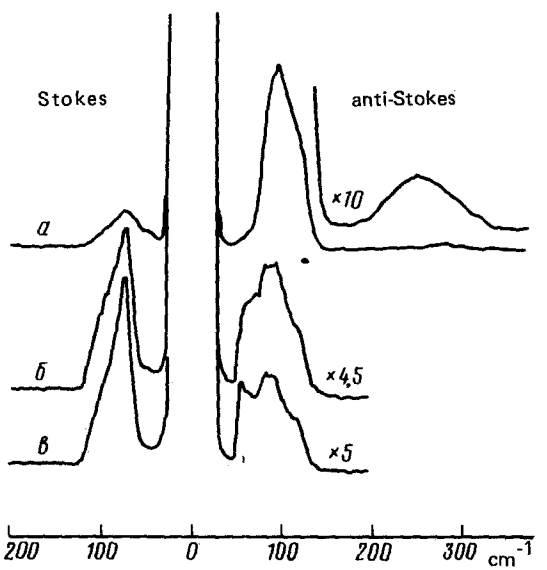

FIG. 1. Spectrograms of the Raman scattering of xenon atoms adsorbed in porous glass. The resolution is 2 cm^{-1}; the recording rate is 15 cm^{-1}/min; the time constant is 2 *s*; and the laser wavelength is 488 nm. a–Spectrum recorded after laser bombardment for 10 h; b–after bombardment for 20 h; c–after bombardment for 30 h.

ponents returned to its original value.

The observed bands can be interpreted as evidence of transitions between different vibrational states of xenon atoms in the adsorption potential. If we assume that the field of the adsorption forces can be described by a Morse potential, we can use these spectra to determine the parameters of the vibrational motion. It turns out that the anharmonicity is $\chi_e\omega_e = 2.7$ cm^{-1}, the depth of the adsorption potential is $D_0 = 1900$ cm^{-1}, the characteristic dimension of the potential is $l = 0.22$ Å, and the fundamental vibrational frequency is $\omega_0 = 143.5$ cm^{-1}. This frequency was determined only within an uncertainty of one or two times the anharmonicity, because of a difficulty in establishing the beginning of the vibrational band corresponding to the 1–0 transition. The second harmonic of the anti-Stokes vibrational band is observed in the frequency interval 200-300 cm^{-1} (spectrogram a). The vibrational structure of the Stokes band is poorly defined.

The comparative intensity of the anti-Stokes band (in comparison with the Stokes band) requires further study, both experimental and theoretical. The same is true of the slow equalization of the intensities of the Stokes and anti-Stokes scattering bands during prolonged laser illumination.

In summary, the interaction of atoms with surfaces can be studied by the effective method of laser Raman scattering by using fine-pore structures which are transparent to the laser beam and by using inert gases, which do not undergo chemical reactions and whose atoms do not exhibit Raman scattering unless they are interacting with a surface.

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