Selective evaporation of frozen gases by laser radiation

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We report observation of selective evaporation, under the influence of a resonant CO_2 cw laser, of the molecules $^{10}BCl_3$ and $^{11}BCl_3$ from a solid film obtained by a freezing boron trichloride at liquid nitrogen temperature on an ir substrate.

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When a laser acts resonantly on a multiphase heterogeneous system, selective heterogeneous adsorption[1] and filtration diffusion[2] processes are observed, as well as a nonthermal shift of the extraction equilibrium.[3] These processes are of interest both from the point of view of studying the singularities of the interaction of vibrationally excited molecules in heterophase systems, and for IR laser photochemistry and laser separation of isotopes.[4]

We report here observation of selective evaporation of frozen gases $^{10}BCl_3$ and $^{11}BCl_3$ under the influence of a CO_2 laser at resonance with the ν_3 vibration of the $^{11}BCl_3$ molecule. Boron trichloride gas with natural isotope content was frozen in the form of a thin film on an IR transparent substrate having the temperature of liquid nitrogen. It turned out that the isotopic structure of the ν_3 band in the solid film of the boron trichloride did not differ noticeably from the structure of the spectrum of the gaseous BCl_3. Freezing causes an equal shift

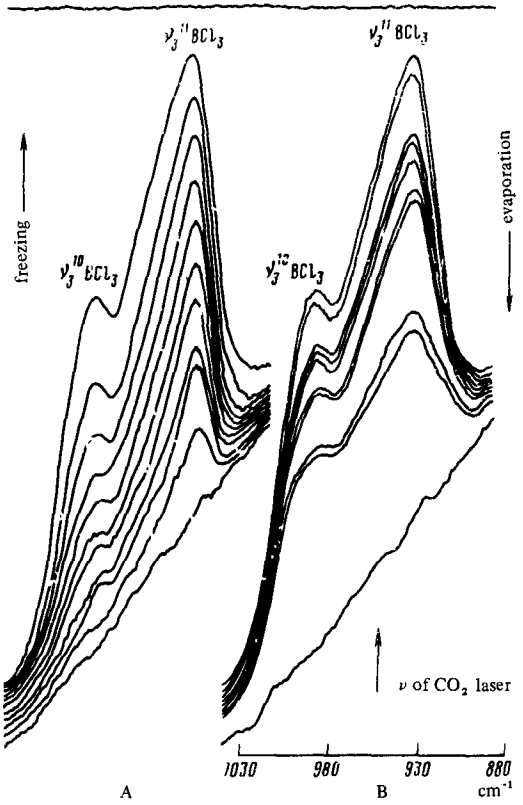

FIG. 1. a) Absorption spectra of ν_3 vibrations in a solid BCl_3 film. The film thickness increases in the upward direction; b) absorption spectra of solid BCl_3 film after laser irradiation. The number of irradiation acts increases in the downward direction.

of the ν_3 bands of the molecules $^{10}BCl_3$ and $^{11}BCl_3$ towards lower frequencies by approximately 20 cm^{-1}, and a small broadening of the bands. Figure 1(a) shows the increase of the absorption in the course of freezing of the BCl_3 on a ZnSe substrate. Estimate based on the absorption coefficient yields for the density of the molecules in the film a value 10^{18} cm^{-2}, corresponding to 100—500 molecular layers. The preservation of the spectral structure after freezing makes it possible to act in resonant fashion on the $^{11}BCl_3$ or $^{10}BCl_3$ molecules in the solid phase.

When the film was exposed to the laser radiation, a predominant evaporation of the resonantly excited $^{11}BCl_3$ molecules was observed, provided that the irradiation was of relatively short duration and of low intensity. Figure 1(b) shows the gradual enrichment of the film with $^{10}BCl_3$ molecules as the number of irradiation acts is increased. The irradiation was by pulses of duration 1—3 min at an intensity 1 W/cm^2. Figure 2 shows the dependence of the enrichment coefficients of BCl_3 in the gas and solid phases as a function of the intensity of the radiation passing through the film. By definition, the enrich-

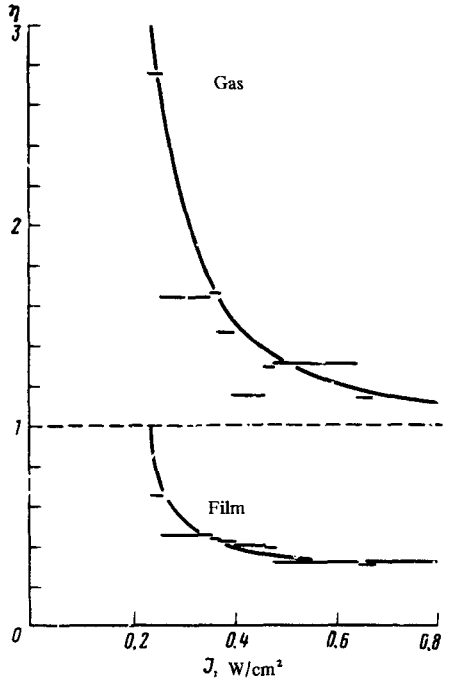

FIG. 2. Dependence of the enrichment coefficient in the gas phase (upper curve) and in the solid film (Lower curve) on the intensity of the laser radiation passing through the film.

ment coefficient is equal to the $^{11}BCl_3/^{10}BCl_3$ concentration ratio after the irradiation, divided by the concentration ratio in the initial gas.

Thus, selective evaporation of $^{11}BCl_3$, with a large enrichment coefficient, is clearly observed. With increasing temperature of the film and substrate, the selectivity of the evaporation decreases. Figure 3 shows the time dependence of the enrichment coefficient in the gas phase, measured with a mass spectrometer against elemental ^{10}B and ^{11}B. Loss of the selectivity of the vibrational excitation of $^{11}BCl_3$ leads to a preferred evaporation of $^{10}BCl_3$.

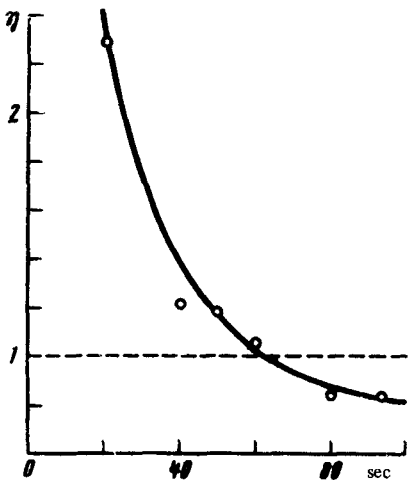

FIG. 3. Dependence of the enrichment coefficient in the gas phase on the irradiation time.

Under the experimental conditions, the enrichment coefficient of the evaporated gas turned out to be directly proportional to the relative concentration of the molecules $^{11}BCl_3/^{10}BCl_3$ in the film before the start of the irradiation, and inversely proportional to the energy consumed in heating the substrate.

In heterogeneous selective processes, the critical energy of the interaction is low. Therefore the absorption of one infrared quantum by the molecule greatly changes the character of the interaction. With the selectivity of the vibrational excitation conserved, a resonant molecule is activated, and in effective vibrational exchange between molecules of different isotopic composition, molecules having large vibrational energy are activated. The results of experiments on selective evaporation of frozen gases by resonant laser radiation agrees with these concepts. In resonant evaporation of molecules quenched in matrices one should expect an increase in the selectivity of the process.

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