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FTIR spectra of liquid argon/liquid nitrogen mixtures: evidence for the existence of a 1:1 bonded species $Ar \cdot N_2$

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Abstract

The mid-infrared spectra of liquid phase argon/nitrogen (80–121 K), krypton/nitrogen (78–121 K) and xenon/nitrogen (80–123 K) mixtures are reported. Superposed on the broad, collision-induced N_2 band, for the Ar/N_2 and the Kr/N_2 mixtures a weak band, proving the existence of a 1:1 species $Ar \cdot N_2$ or $Kr \cdot N_2$, was observed near 2326 cm^{-1}. Using spectra recorded at different temperatures, the complexation enthalpy of $Ar \cdot N_2$ was determined to be $-1.5(4)$ kJ mol^{-1}.

1. Introduction

For some time, we have been using solutions in liquid argon and in liquid nitrogen for the study of weakly bound molecular complexes [1–5]. To obtain a more detailed understanding of the solvation influences on the complexes, we have recently initiated a study in mixtures of liquid argon and liquid nitrogen [6,7]. For the mixed solvents, in the infrared (IR) spectra a weak, narrow band was observed on the low-frequency side of the broad, collision-induced N_2 stretching band, at 2326 cm^{-1}. This band was observed even in the absence of a solute but could not be detected in the spectra of pure liquid argon or of pure liquid nitrogen. This suggests that the band is due to a species formed between Ar and N_2, for which a 1:1 Van der Waals complex is the primary candidate. Such complexes between nitrogen and the rare-gas atoms have drawn substantial attention [8–24]. The complex between Ar and N_2, for example, was first observed by Henderson and Ewing [9] using low-resolution (1 cm^{-1}) gas-phase IR spectroscopy. Later, the available data were expanded using high-resolution FTIR [14], FT microwave spectroscopy [16,17,21], scattering experiments [19,22] and ab initio calculations [20,23,24]. As a consequence, the structure of the complex and the potential governing the interaction between nitrogen and argon are characterised rather well.

The present study is dedicated to finding evidence for the origin of the 2326 cm^{-1} band in an Ar/N_2 complex. Therefore, in this Letter, we report on the IR spectra of liquid mixtures of argon and nitrogen, obtained at different temperatures and different Ar/N_2 concentration ratios. It will be shown that the band indeed arises in a 1:1 complex, and a value for the complexation enthalpy will be derived. In addi-

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