Influence of the nanodroplet size, interaction potential and vibrational energy gap on the vibrational relaxation of diatomic molecules in superfluid helium nanodroplets

Miquel Blanchafort-Jorquera, Arnau Vilà and Miguel González

Departament de Ciència dels Materials i Química Física and IQTC, Universitat de Barcelona. C/ Martí i Franquès 1, 08028 Barcelona. miguel.gonzalez@ub.edu

1. INTRODUCTION

The study of reactions and energy transfer processes involving 4He superfluid nanodroplets [HeNDs (quantum fluid); T=0.37 K] and atoms or molecules is gaining progressively more interest in Chemistry.1-2 The vibrational energy relaxation (VER) of a homonuclear diatomic molecule in a 4He superfluid nanodroplet (HeND) was studied using a hybrid quantum approach proposed by us.3-4,5 This work extends a previous contribution of our own on the VER of the 1_J(X) molecule in HeNDs,6 and corresponds to the second theoretical investigation reported so far on this interesting problem. The VER from v=1 to v=0 of 1_J(X) embedded in a 100 4He atoms nanodroplet has been taken as reference system and a number of situations has been simulated: (1) number of 4He atoms (50-200); (2) "He-1_J" interaction potential energy from 0.5 to 1.5 times the real He-1_J interaction; (3) 1_J (or X2) v:0-1 vibrational energy gap from 0.75 to 1.5 times the real 1_J energy gap.

2. THEORETICAL METHOD

Hybrid approach

• 4He : Phenomenological TDDFT (Time Dependent Density Functional Theory) using the Orsay-Trento functional7

• X2 : Standard time dependent quantum mechanics (wave function)

\[ i\hbar \frac{\partial}{\partial t} \psi_{X_2}(R_{He}) = -\frac{\hbar^2}{2m_{X_2}} \nabla^2 + \int dr V_{He-X_2}(r, R_{He}) \psi_{X_2}(r) + |\psi_{He}(R_{He})| \]

3. RESULTS AND DISCUSSION

3.1. Vib. relaxation and nanodroplet size 1_J@4He

3.2. Vib. relaxation and interaction potential energy X2@4He

3.3. Vib. relaxation and vibrational energy gap X2@4He

4. CONCLUSIONS

The HeND size has a little effect on the VER dynamics, as the interaction between the molecule and the liquid helium mainly comes from the first solvation shell (which is fully formed for all the HeND sizes investigated).

The interaction potential energy and the v:0-1 energy gap have an important influence on the VER time properties, which decrease in a significant way as the former becomes stronger and the second decreases.

The molecule-helium interaction energy is hardly modified during the relaxation process, but it shows oscillations during the transition. Thus, the HeND structure is essentially unchanged.

An interpretation based on the coupling terms, \( <V_i|^2 > \), between the two vibrational levels involved and the oscillator velocity (that is proportional to \( \nu_i^{(2)} \)) has been attempted.

REFERENCES


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