

## SUPPORTING INFORMATION

### Vibrational energy relaxation of a diatomic molecule in a superfluid helium nanodroplet. Influence of the nanodroplet size, interaction energy and energy gap

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**Table s1.** Energy ( $E$ ) contributions (in K) at  $t=0$  for  $I_2(\nu=1)@(^4\text{He})_N$ , as a function of  $N$ <sup>a</sup>

$N$	$E(\text{He})_N$	$E$ per atom $(\text{He})_N$	Kinetic $E(\text{He})_N$	Lennard-Jones $E(\text{He})_N$
50	99.73	1.99	185.39	-141.26
100	-102.45	-1.02	214.45	-403.94
150	-338.14	-2.25	230.49	-674.32
200	-588.94	-2.94	241.10	-952.21

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$N$	Interaction $E$			Total $E I_2@(\text{He})_N$
	Correlation $E(\text{He})_N$	$I_2-(\text{He})_N$	Vibrational $E(I_2)$	
50	55.30	-664.09	459.96	-104.40
100	86.45	-739.99	459.96	-382.48
150	104.98	-767.58	459.96	-645.76
200	121.41	-782.70	459.96	-911.68

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<sup>a</sup>  $E(\text{He})_N$  refers exclusively to the helium and includes several terms: kinetic energy, potential energy (Lennard-Jones potential energy function) and correlation energy.

**Table s2.** Inverse of the global relaxation times, lifetimes and transition times of the  $\nu=1$  excited state and  $\langle V_{01}^2 \rangle$  coupling terms for the five interaction potential energies considered ( $V=xV_{I_2-He}$ ) and  $N=100$ .<sup>a</sup>

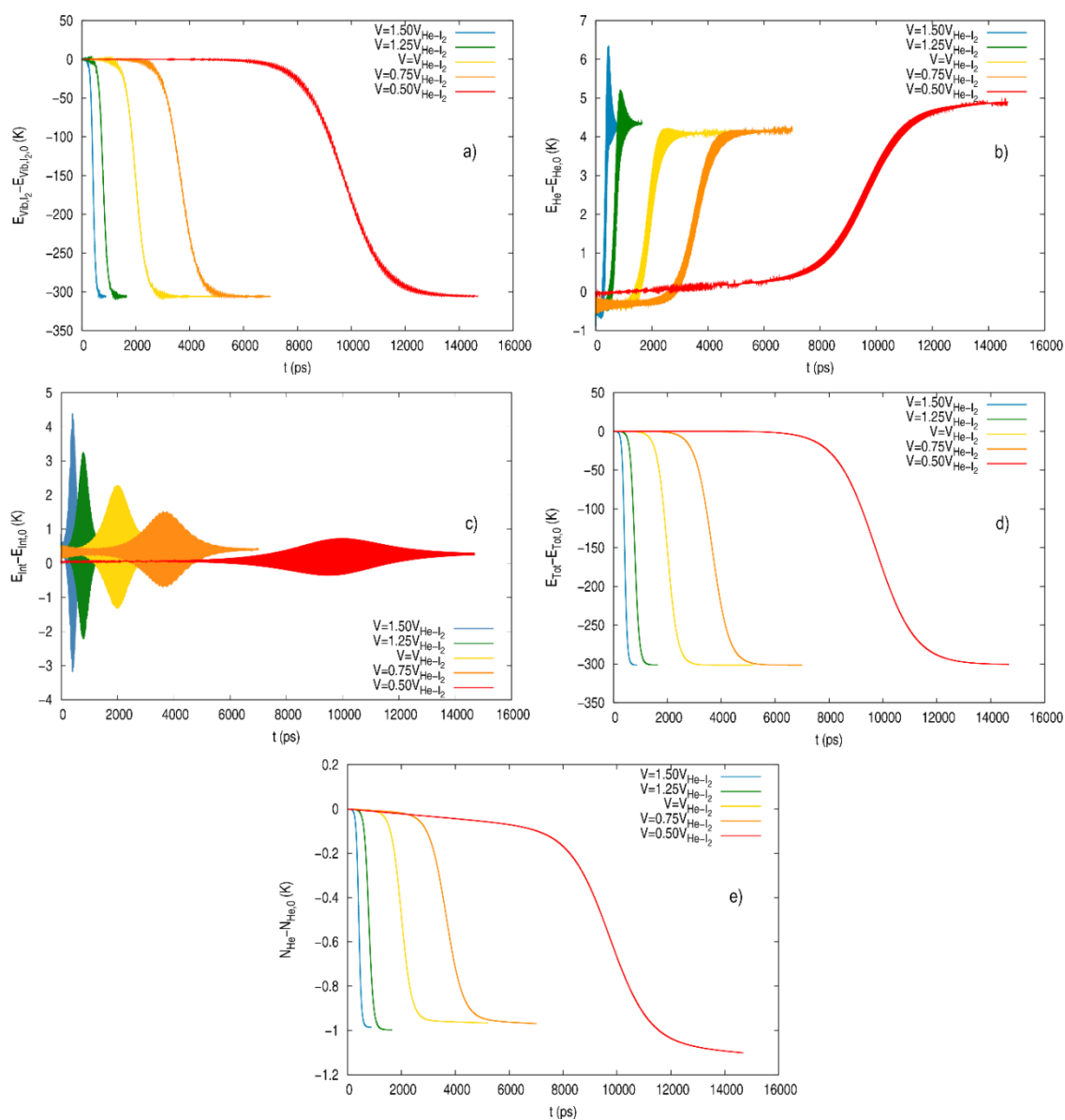
$x$	$\langle V_{01}^2 \rangle$ (K <sup>2</sup> )	1/Global relax. time (ps <sup>-1</sup> )	1/Lifetime (ps <sup>-1</sup> )	1/Transition time (ps <sup>-1</sup> )
0.50	0.15 (0.09)	$7.66 \cdot 10^{-5}$ (0.22)	$1.03 \cdot 10^{-4}$ (0.21)	$1.49 \cdot 10^{-4}$ (0.26)
0.75	0.61 (0.39)	$1.96 \cdot 10^{-4}$ (0.56)	$2.73 \cdot 10^{-4}$ (0.54)	$3.50 \cdot 10^{-4}$ (0.60)
1.00	1.56 (1.00)	$3.48 \cdot 10^{-4}$ (1.00)	$5.01 \cdot 10^{-4}$ (1.00)	$5.79 \cdot 10^{-4}$ (1.00)
1.25	3.09 (1.98)	$8.50 \cdot 10^{-4}$ (2.44)	$1.27 \cdot 10^{-3}$ (2.53)	$1.34 \cdot 10^{-3}$ (2.31)
1.50	5.98 (3.84)	$1.56 \cdot 10^{-3}$ (4.49)	$2.42 \cdot 10^{-3}$ (4.83)	$2.31 \cdot 10^{-3}$ (4.00)

<sup>a</sup>  $\langle V_{01}^2 \rangle$  and the inverse of the relaxation times relative to the  $x=1.00$  reference case are given between parentheses.

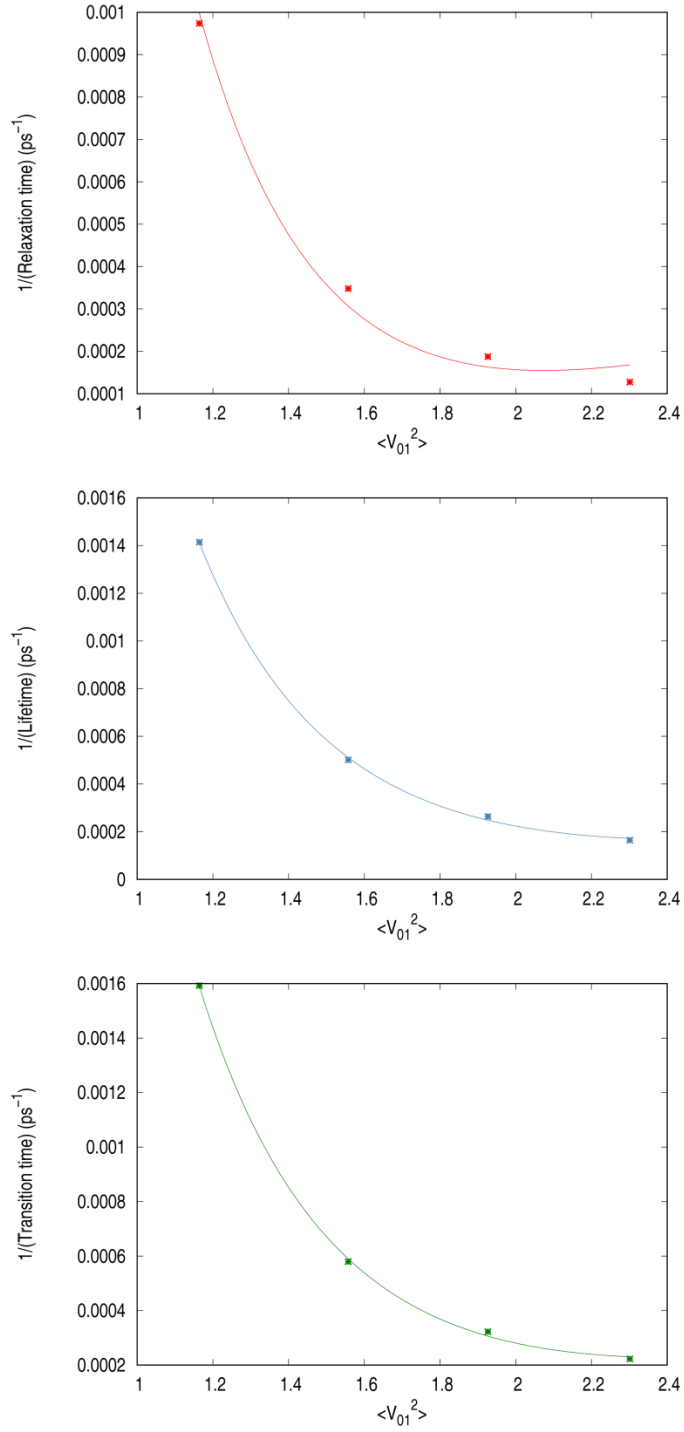
**Table s3.** Inverse of the global relaxation times, lifetimes and transition times of the  $\nu=1$  excited state,  $\langle V_{01}^2 \rangle$  coupling terms and  $\sqrt{x}$  for the four vibrational frequencies  $\nu_e$  considered ( $\nu_e = x\nu_{e,I_2}$ ) and  $N=100$ .<sup>a</sup>

$x$	$\sqrt{x}$	$\langle V_{01}^2 \rangle$ (K <sup>2</sup> )	1/Global relax. time (ps <sup>-1</sup> )	1/Lifetime (ps <sup>-1</sup> )	1/Transition time (ps <sup>-1</sup> )
0.75	0.87	1.16 (0.75)	$9.74 \cdot 10^{-4}$ (2.80)	$1.41 \cdot 10^{-3}$ (2.82)	$1.59 \cdot 10^{-3}$ (2.75)
1.00	1.00	1.56 (1.00)	$3.48 \cdot 10^{-4}$ (1.00)	$5.01 \cdot 10^{-4}$ (1.00)	$5.79 \cdot 10^{-4}$ (1.00)
1.25	1.12	1.93 (1.24)	$1.87 \cdot 10^{-4}$ (0.54)	$2.64 \cdot 10^{-4}$ (0.53)	$3.22 \cdot 10^{-4}$ (0.56)
1.50	1.22	2.30 (1.48)	$1.28 \cdot 10^{-4}$ (0.37)	$1.65 \cdot 10^{-4}$ (0.33)	$2.23 \cdot 10^{-4}$ (0.38)

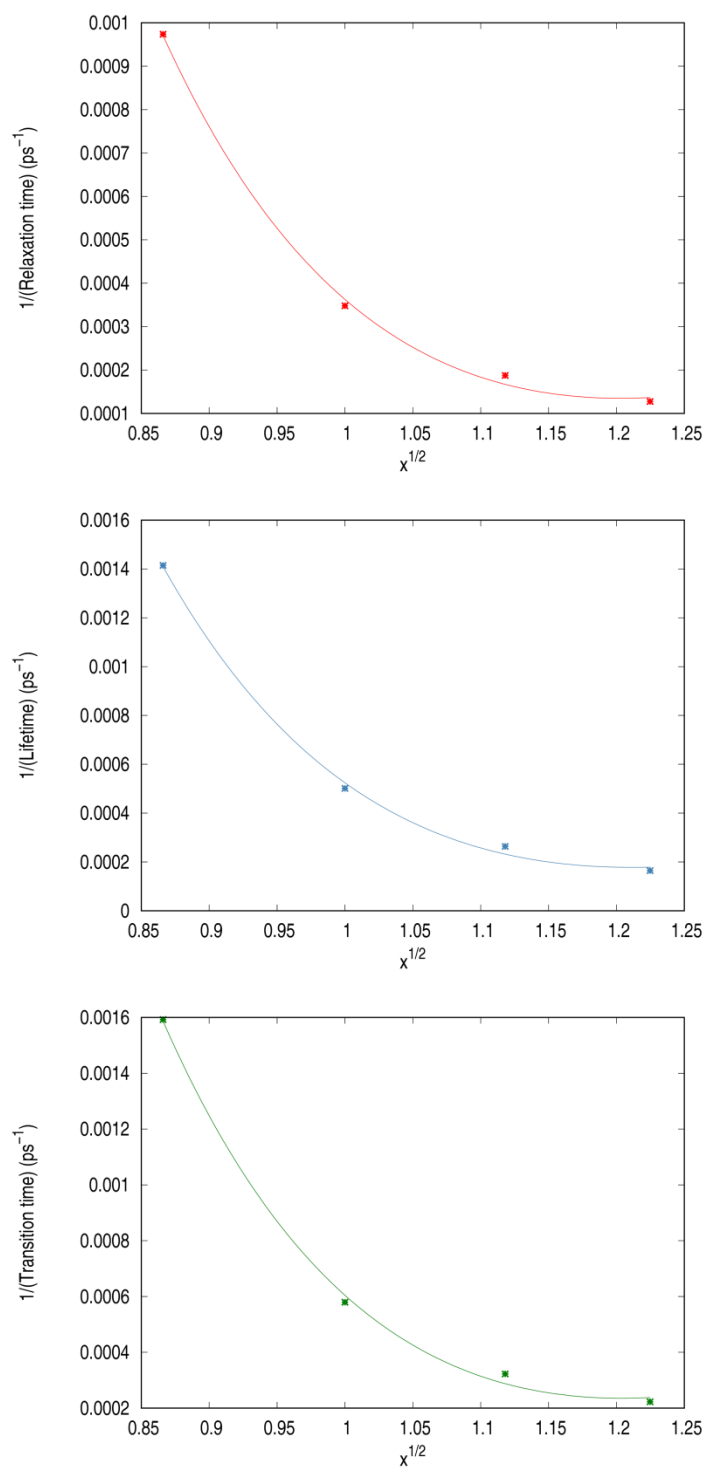
<sup>a</sup>  $\langle V_{01}^2 \rangle$  and the inverse of the relaxation times relative to the  $x=1.00$  reference case are given between parentheses.



**Figure s1.** Energy increments involved in the VER from  $\nu=1$  for the five selected “I<sub>2</sub>-HeND” interaction potential energies and number of  $^4\text{He}$  atoms of the nanodroplet, as a function of time for  $N=100$ : a) vibrational energy of the molecule; b) helium energy of the nanodroplet; c) molecule-nanodroplet interaction energy; d) total energy of the system (I<sub>2</sub>@HeND); e) number of  $^4\text{He}$  atoms of the HeND.



**Figure s2.** Inverse of the global relaxation time (top), lifetime (middle) and transition time (bottom) of the  $\nu=1$  excited state, as a function of the  $\langle V_{01}^2 \rangle$  coupling term for the vibrational frequencies selected ( $\nu_e = x\nu_{e,I_2}$ , with  $x=0.75, 1.00, 1.25$  and  $1.50$ ) and  $N=100$ . The coupling term increases with  $x$ .



**Figure s3.** Inverse of the global relaxation time (top), lifetime (middle) and transition time (bottom) of the  $\nu=1$  excited state, as a function of the  $x^{1/2}$  term for the vibrational frequencies selected ( $\nu_e = x\nu_{e,I_2}$ , with  $x=0.75, 1.00, 1.25$  and  $1.50$ ) and  $N=100$ .