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Influence of complexation and solid environment on the vibrational coherence of DCl

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Abstract. The vibrational dynamics of DCl molecules embedded in different cryogenic matrices is studied by time resolved infrared Degenerate Four Wave Mixing experiments using the infrared free electron laser CLIO. The coherence behaviour of the D−Cl stretch in pure argon as well as in mixed argon/nitrogen van der Waals solids is investigated. Different interactions between the excited molecules and their environment are probed: the van der Waals interaction between the trapped molecules and the solid lattice and the interaction binding complexes trapped in the matrix (either the vdW interaction between DCl and nitrogen or weak H bonds in $(DCl)_n$ clusters). The dependence of the dephasing time on the solid and on specific interactions is clearly observed.

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1 Introduction

The knowledge of molecular vibrational dynamics in the electronic ground state is essential in the understanding of numerous physical or chemical processes, especially in the condensed phase [1]. The dynamics of an oscillator is very sensitive to its environment and differs widely from a system to another when going from the gas phase to liquids and solids. In principle, information on environmental effects on the vibrational dynamics can be directly obtained from the analysis of the homogeneous absorption lines. However, the broadening of the spectral lines in condensed phase is often dominated by inhomogeneous effects arising from static interactions of the oscillator with its surrounding. In such cases, non-linear experiments are required to extract the homogeneous contribution hidden by inhomogeneous broadening [2]. Time resolved one-color Degenerate Four Wave Mixing (DFMW) techniques are powerful tools to access the vibrational dynamics: the population relaxation time T_1 (from pump-probe experiments), the coherence time T_2 (from photon echoes measurements) and the "pure" dephasing time T_2^*, very sensitive to the molecular surroundings [3,4], related to the previous by the following equation:

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}. \tag{1}$$

In order to determine T_2, time resolved experiments have been performed by means of a 2-pulses photon echoes experimental set-up using the infrared Free Electron Laser CLIO (Centre Laser Infrarouge d'Orsay), (see Sect. 2.1). The peculiar temporal structure of the FEL emission allows the observation of accumulated photon echoes on molecular systems with long vibrational relaxation times, such as small molecules isolated in cryogenic matrices [4]. In this last case, in addition to the measurement of dephasing times, our experiment gives access to another important dynamical process, the spectral diffusion.

Hydrogen chloride molecules embedded in van der Waals solids are a very interesting system because both isolated molecules and small sized clusters involving hydrogen bonding are present in the solid. In practice, we use DCl instead of HCl because the absorption wavelength of the deuterated species ($\lambda = 4.8$ μm) matches to the spectral range of the FEL better than the HCl wavelength ($\lambda = 3.5$ μm) and also because this spectral region is free from atmospheric absorption. Particular attention is paid to the dimers in which the two H(D)Cl molecules are differently perturbed by the hydrogen bond [5,6]; the two H(D)−Cl stretching modes (ν_1 and ν_2) have their frequencies down shifted from the monomer frequency, the frequency of ν_2 (the "bonded" mode) corresponding to the

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stretch directly involved in the hydrogen bond is more red shifted than that of ν_1 (the "free" mode) corresponding to the less perturbed stretch. We investigate rare gas matrices (solid argon) and mixed nitrogen/rare gas matrices. Mixed matrices may be used in order to study molecular complexes with DCl molecules: in mixed N_2/Ar matrices including a small amount of nitrogen, complexes with nitrogen are easily isolated [7]. DCl molecules occupy a single substitutional site of the N_2 and Ar f.c.c.lattices, and thus of the mixed N_2/Ar matrix. In solid argon, the rotation of isolated DCl is spectrally observed, but the complexation, even with nitrogen, prevents the rotational structure [7]. Depending on the DCl concentration, several lines arising from dimers, trimers and higher clusters appear in the same sample. Depending on the N_2 concentration in argon matrices, nitrogen complexes of different sizes are observed.

From previous works in matrices [8], the vibrational population relaxation time T_1 of the isolated molecule is known to be very long, in the microsecond range. For the dimer species, only estimations from the gas phase are available [9]. Nevertheless, in most cases, by distinguishing between two-pulse photon echo signals and accumulated photon echo signals, one obtains information on the order of magnitude of T_1 [4,10,11]: if T_1 is longer than 16 ns (a time fixed by the time profile of the FEL emission) accumulated photon echoes are observed. In such case the measured T_2 time is equal to the "pure" dephasing time (see Eq. (1)). Moreover, the spectral diffusion, due to intermolecular vibrational energy transfers between the guest molecules (V-V transfer), can be studied.

This paper presents a study on the influence of the environment on the vibrational coherence of isolated and clusterized DCl molecules in argon and mixed nitrogen/argon matrices. The experimental results, particularly the vibrational dephasing and the V-V transfer processes are compared with those obtained in nitrogen matrices [10,12]. The influence of the solid and complexation effects on the vibrational dephasing processes are highlighted. The experimental methods and the general features of the non-linear response are summarized in Section 2. Section 3 describes the results obtained in cryogenic samples containing DCl and nitrogen molecules trapped in solid argon. Finally, in Section 4 the discussion and the comparison between the different $(DCl)_n(N_2)_x(Ar)_y$ systems are presented.

2 Methods

2.1 Experimental

Experiments were carried out in a liquid helium bath cryostat (Air Liquide) working down to 7 K. The gas mixture DCl/HCl/N_2/Ar was deposited on a cold sapphire window maintained at 20 K. DCl was obtained from Merck Sharp Dohme Isotopes. The variable adsorption of the H(D)Cl on the stainless steel vacuum manifold makes exact concentration determination difficult, and hydrogen deuterium exchange on the walls leads to a complex absorption spectra. The final composition of the solid samples was checked and analyzed by measuring absorption spectra by means of a Mattson FTIR spectrometer with a resolution of 0.25 cm^{-1}.

The energy of the photons delivered by the FEL [13, 14] was tuned between 2030 and 2070 cm^{-1} with a typical laser spectral width around 15 cm^{-1}. This spectral resolution enables a selective excitation of the monomers, dimers, complexes and large clusters. The FEL has a peculiar time sequence: sub-picosecond pulses are supplied at a rate of 62.5 MHz in 10 μs long bunches, and the repetition rate of bunches is 25 Hz.

The DFWM experimental set-up has been described in details elsewhere [3]. Briefly, the laser beam was splitted in two beams. The first one is sent directly onto the sample, while the second one is delayed. This is thus a two-pulse photon echoes experiment, a particular case of three-pulse photon echoes, where the waiting time T (the delay between the two last pulses) is set to be zero, and where the two last pulses have the same wave vector $\mathbf{k}_2$. At the exit of the sample, the DFWM signal was emitted in a well defined direction $\mathbf{k}_s = 2\mathbf{k}_2 - \mathbf{k}_1$ where $\mathbf{k}_{1,2}$ are the directions of the two laser beams crossed in the sample, $\mathbf{k}_2$ corresponding to the beam delayed by τ from the other. The signal, geometrically selected by a well-positioned iris, is detected onto a liquid nitrogen cooled MCT detector, integrated over the 10 μs bunch and recorded by the computer. The zero delay time ($\tau = 0$) was determined from experiments on $W(CO)_6$ in CCl_4 solution performed with the same geometry of the laser beams before the experiments in matrices [15]. The temporal resolution of our experiments limited by the laser pulsewidth can be modified in the range of [0.6, 1.5 ps] by adjusting the laser cavity length [16].

2.2 Description of the non linear responses

As mentioned above, our experimental set-up allows the observation of either two pulse photon echo signals, or stimulated photon echo signals. In most cases, the distinction between the two kinds of signals is clearly observed in the negative part of the signal ($\tau < 0$): in the case of a two-photon echo, the signal just reflects the temporal width of the laser pulse (~1 ps), whereas in the other case, the signal is related to the Fourier Transform of the absorption profile. Thus, a look on this negative part gives a clear indication on the population relaxation time of the probed vibrational modes: this time must be longer than $T_R = 16$ ns, T_R is the repetition time of FEL pulses in a bunch, when stimulated (i.e. accumulated) photon echoes are observed, and in this case, $T = T_R$ is the fixed time delay between the two last pulses of the three-pulse sequence of the stimulated echo [4]. Most of the detected signals $S(\tau)$ described here are stimulated accumulated three pulse photon echo signals. The shapes of the signals are discussed in references [10,12,17]. We report here only general tendencies.

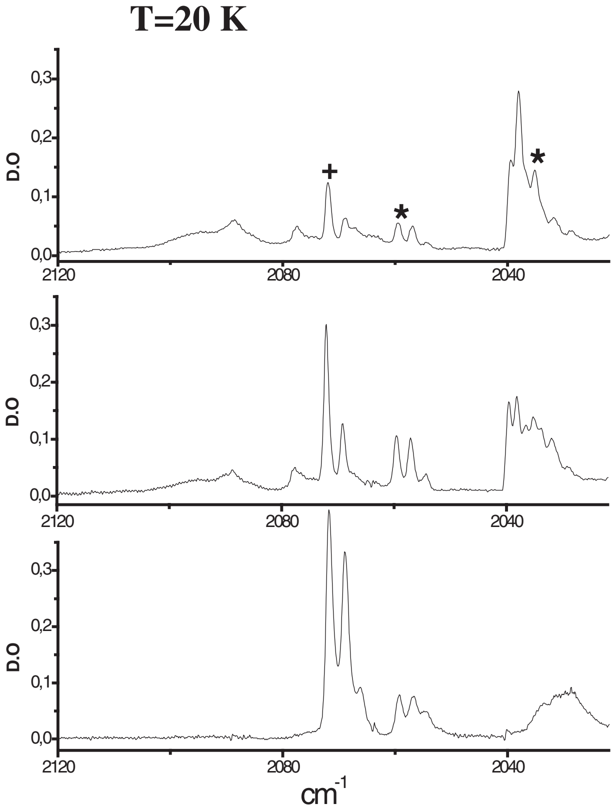

Fig. 1. Absorption spectra of DCl in solid argon with nitrogen impurities at 20 K. The concentration of nitrogen increases from the upper to the bottom spectrum. The position of the two stretching modes (ν_1 and ν_2) of the $(DCl)_2-(N_2)$ complexes is marked by an asterisk. The sign + indicates the $DCl-N_2$ species.

Table 1. Main vibrational transition frequencies of $D^{35}Cl$ and its dimers in mixed N_2/Ar matrix ($T = 20$ K); the corresponding absorption bands assigned to the $D^{37}Cl$ isotopic species are redshifted from these values by 2.8−3 cm^{-1}, depending on the band.

wavenumber (cm^{-1})	assignment [7,22,23] (cm^{-1})
2094.8	R(1) monomer
2089.5	R(0) monomer
2078.5	Q monomer
2072.9	$DCl-N_2$
2069.9	$DCl-(N_2)_2$
2069.5	ν_1 dimer, very weak
2068.6	P(1) monomer
2060.4	ν_1 dimer-N_2
2060.3	ν_1 DCl−HCl dimer-N_2
2057.9	ν_1 dimer-N_2
2057.8	ν_1 DCl−HCl dimer-N_2
2040.5	ν_2 HCl−DCl dimer
2039.0	ν_2 dimer
2034.5	ν_2 dimer-N_2
2032.8	ν_2 dimer-N_2

When spectral diffusion occurs, the DFWM signals can exhibit two components: a short and a long one. The former is the signature of efficient V-V transfers (spectral diffusion), and the latter involves the coherence time T_2 of the excited transitions, which is coming from the rephasing process [4,18]. The relative weight of the two components allows the estimation of V-V transfer rates in the sample [12].

Because of its spectral width, the laser source can also coherently excite transitions of different species. In such situation, if $P_i^{(3)}(t,\tau)$ is the component of the third order polarization of species i in direction $\mathbf{k}_s$, the DFWM signal is:

$$S(\tau) = \int_0^{\infty} |\sum_i P_i^{(3)}(t,\tau)|^2 dt, \qquad (2)$$

where the time origin is the time of the interaction with the third laser pulse. Therefore, $S(\tau)$ includes the non linear response of all excited transitions and does not have a simple theoretical expression. The coherent excitation of several transitions is responsible for the oscillatory behaviour of the signal, sometimes observed especially in the short component [10,12,17,19]. We have built a simplified analytic model to describe the signal, taking into account the spectral diffusion and the simultaneous excitation of different transitions [10,12]. If the long component of $S(\tau)$ is too weak, the observed signal is fitted using an expression deduced from the model, involving dephasing times T_2 and taking into account the V-V transfers, occurring during the long delay T_R [4,12]. Otherwise, the time T_2 is directly extracted from this long component, which follows a $\exp(-4\tau/T_2)$ exponential decay.

3 Results: DCl in solid argon

The spectrum of isolated HCl/DCl molecules in argon is characterized by the presence of the rotation lines R(0), R(1), Q and P(1), evidencing the quasi free rotation of the hydrogen halide molecules in the argon environment. A fast dephasing process is expected because of this rotation and its efficient coupling to the lattice phonons [20]. As a matter of fact, a very short time-resolved signal is observed, no longer than few picoseconds. The study of the dynamical behaviour of DCl monomers rotating in solid argon will not be discussed here. Due to the rotation, a comparison between the vibrational dephasing times for isolated DCl in solid argon and in solid nitrogen where no rotation occurs [21] does not make sense.

Our attention is focused on nitrogen DCl complexes and DCl clusters embedded in solid argon. The absorption spectra revealed the presence of HCl/DCl complexes with nitrogen in the solid argon matrix even in the case of a very weak amount of nitrogen. Depending on the N_2 concentration, characteristic bands assigned to polymeric species $(H(D)Cl)_x-(N_2)_n$ are observed (Fig. 1, Tab. 1). The strong affinity of hydrogen halide to form complexes with nitrogen make inevitable this issue. The concentration of residual nitrogen could only be estimated from a comparison between our data with those obtained

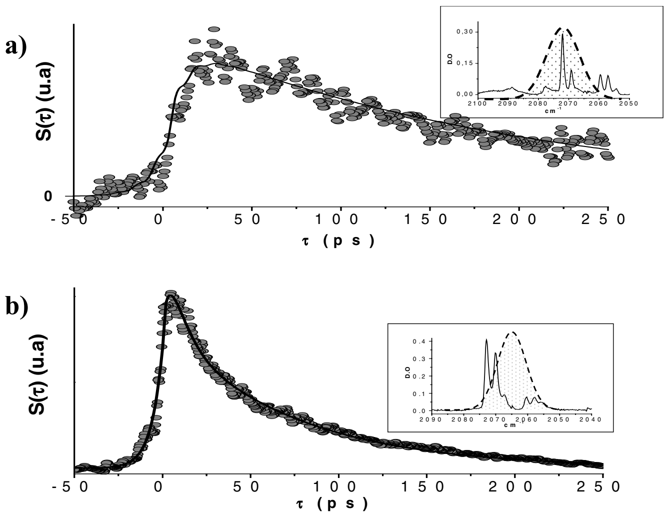

Fig. 2. DFWM signals at 6 K of the DCl$-$(N$_2$)$_x$ complexes. (a) $x = 1$, (b) $x = 2$. The solid line corresponds to the fit obtained on the whole signal following the model presented in Section 2.2. In the inserts, the spectral position of the laser excitation is represented.

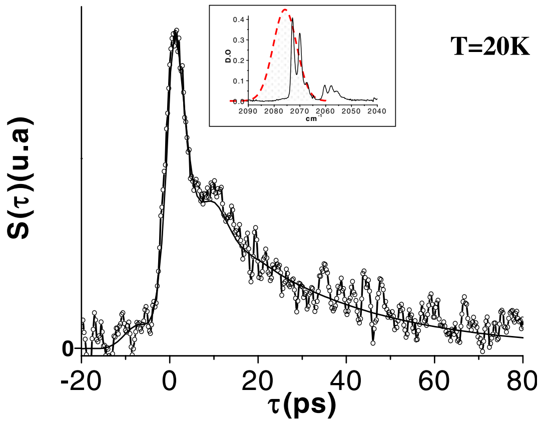

Fig. 3. DFWM signal at 20 K of the DCl$-$(N$_2$) complex in a sample containing a few percent of nitrogen. A short component involving oscillations is observed, superimposed to a long component ($T_2 = 150$ ps). The solid line corresponds to the fit obtained on the whole signal following the model presented in Section 2.2. In the insert, the spectral position of the laser excitation is represented.

in mixed matrices by the Perchard's group [7,22]. From top to bottom of Figure 1, the concentration of nitrogen (c_{N_2}) is evaluated to $c_{\mathrm{N}_2} < 0.2\%$, $0.2\% < c_{\mathrm{N}_2} < 0.4\%$ and $2\% < c_{\mathrm{N}_2} < 5\%$ respectively. In the last case, the absorption of isolated DCl molecules is not observed, all DCl molecules are complexed with nitrogen in the sample. The enhanced formation of nitrogenous complexes, as compared to a statistical distribution, is related to DCl$-$N$_2$ interaction which is stronger than the DCl$-$Ar interaction as revealed in the study of the DCl monomer trapped in solid nitrogen through the enhancement of the transition dipole moment and the efficient dephasing process of the isolated DCl molecules [12].

Taking advantage of the FEL tunability, we have performed time resolved measurement on the bands assigned to these (D(H)Cl)$_x-$(N$_2$)$_n$ complexes.

3.1 DCl complexed with one or two nitrogen molecules

The bands in the range [2067$-$2073 cm^{-1}] are assigned to complexes between DCl (^{35}Cl or ^{37}Cl isotopic species) and one or two nitrogen molecules (Tab. 1). DFWM signals correspond to stimulated accumulated photon echo signals, with long or very long decays. Moreover, the short component related to spectral diffusion can only be distinguished in samples including a few percent of nitrogen. In most cases, echo decay curves are not far from a monoexponential shape (Figs. 2 and 3).

DFWM signals of DCl$-$N$_2$ complexes are observed either for a very weak nitrogen concentration or under selective excitation of the DCl$-$N$_2$ band. At the lowest temperatures ($T = 6-8$ K), the coherence time is in the nanosecond range (Fig. 2a): 1.3$\pm$0.6 ns at 6 K, 1.0$\pm$0.4 ns at 7.5 K. It decreases to 0.15$\pm$0.05 ns at $T = 20$ K (Fig. 3).

DCl$-$(N$_2$)$_2$ complexes are studied in samples containing a few percent of nitrogen. A selective excitation cannot be used because of the overlap between D^{37}Cl$-$N$_2$ and D^{35}Cl$-$(N$_2$)$_2$ absorption bands. By tuning the laser frequency at 2067 cm^{-1}, we obtain the signal reproduced in Figure 2b: the DFWM signal shows an exponential decay with characteristic time (0.29 $\pm$ 0.02 ns) faster than that of the DCl$-$N$_2$ complexes, it corresponds obviously to the behaviour of DCl$-$(N$_2$)$_2$ complexes. At low temperature, the DCl$-$(N$_2$)$_2$ coherence time is four times shorter than that of DCl$-$N$_2$ complexes. The temperature dependence of DCl$-$(N$_2$)$_2$ coherence time has not been clearly established in these experiments; at higher temperatures, we have not been able to distinguish the behaviour of DCl$-$N$_2$ and DCl$-$(N$_2$)$_2$ complexes.

Spectral diffusion due to quasi-resonant intermolecular vibrational energy transfer between the trapped species (V-V transfer) is only observed between DCl$-$N$_2$ complexes in the samples where the nitrogen concentration reaches a few percents because the V-V transfer efficiency is strongly dependent on the distances between the probed species, i.e. on their concentration. DFWM signals also show an enhancement of this dynamical process with the temperature. At 20 K, the short component is clearly observed (Fig. 3). It exhibits oscillations due to the coherent excitation of the different isotopic complexes. The analysis of the whole DFWM signal establishes that V-V transfers occur between all these isotopic species, leading to the observation of "quantum beats" [17]. A characteristic time of V-V transfer (T^{VV}) can be extracted from the analysis of the time-resolved signal, following the expression established in reference [12]. This time, characteristic of transfers between nitrogen complexes, is estimated to be 64 ns at 6 K and decreases to 25 ns at 20 K. In addition, assuming that V-V transfers are induced by dipole-dipole interactions, T^{VV} can be calculated using standard approximations (see Ref. [12] for details) from the concentration of the probed species deduced from the absorption spectra, the inhomogeneous width measured on the absorption bands, the homogeneous width measured by the coherence time. A good agreement with the previous values is obtained using this description of V-V transfers when a small enhancement by a factor of about 1.3 of the transition dipole moment of DCl is taken into account in the calculations. Such an enhancement should not be surprising. Indeed the same assumption of the increase of the transition dipole moment of DCl has been made in the study in solid nitrogen, from which an enhancement of a factor of 2 from the gas phase has been deduced. Moreover, such a temperature dependence is expected from the calculations, as described in reference [12], when the homogeneous linewidth is much shorter than the inhomogeneous one, estimated to 1 cm^{-1} in the case under study.

Some conclusions also arise from the results on the coherence times. The observation of long coherence times signifies weak couplings between the vibrational mode and phonon modes. If the temperature dependence of DCl$-$N$_2$ coherence is due to the activation of a phonon mode, this mode would have a low frequency, close to ten wavenumbers, corresponding to the lowest frequencies of the argon lattice phonon band. On the other hand, we observe that when the number of complexed nitrogen molecules increases, the coherence time converges towards the value obtained for the isolated DCl trapped in solid nitrogen ($\sim$175 ps at 5.6 K) [12].

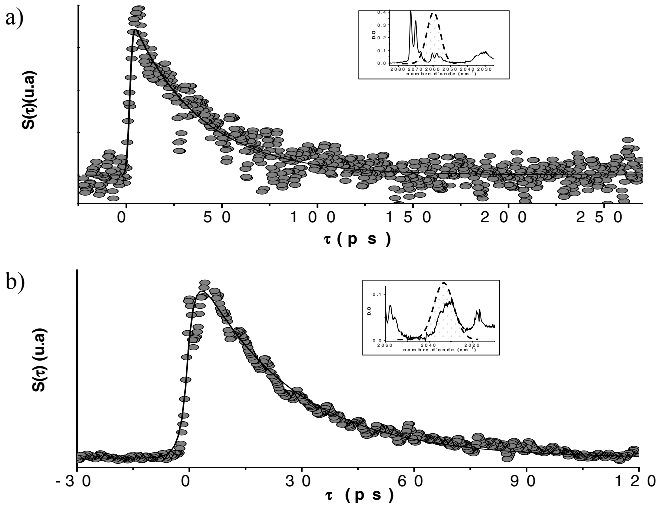

Fig. 4. DFWM signals of the (DCl)$_2-$(N$_2$) complexes from a mixed Ar/N$_2$ matrix where all the guest molecules are complexed with nitrogen. (a) Excitation of the ν_1 mode at 6 K, (b) excitation of the ν_2 mode at 20 K. The solid line corresponds to the fit obtained on the whole signal following the model presented in Section 2.2. In the inserts, the spectral position of the laser excitation is represented.

3.2 DCl dimers complexed with nitrogen

When dimers are embedded in pure argon matrices, the absorption band of the mode ν_1 is nine times less intense than the absorption band of the mode ν_2 [23]. On the contrary, in the case of dimers complexed with a nitrogen molecule, this difference in intensities is much less important, and DFWM signals on both modes are more easily observed.

The shape of the DFWM signals at $\tau < 0$ indicates that all the observed signals on the dimers result from stimulated echoes. Population relaxation times of the dimer vibrational levels are longer than 16 ns for both modes (ν_1 and ν_2). No short component is distinguished in the time evolution, and the signals are analyzed without involving spectral diffusion. The concentration of DCl dimers in the samples of interest is too low to observe the effect of V-V transfers.

With a laser excitation centered around 2055 cm^{-1}, the region assigned to the mode ν_1 of the complexed dimer (DCl)$_2-$N$_2$ is excited. A typical result is shown in Figure 4a. As for isolated DCl molecules complexed with nitrogen, the echo signal decays exponentially and at $T = 6$ K, a dephasing time of about 150 ps is extracted.

By exciting around 2030 cm^{-1}, the ν_2 stretching mode of (DCl)$_2$ and (DCl)$_2-$N$_2$ species are probed. In contrast with the case of mode ν_1, the complexed dimer cannot be selectively excited because of the strong IR activity of the "bonded" mode in both (complexed or isolated) dimers. One can assume that in the samples including a few percents of nitrogen, all the DCl species, monomers and dimers, are complexed with nitrogen. Nevertheless, in any case, from less than 0.2% to few percents of nitrogen in the matrix, shorter dephasing T_2 times are measured (see Fig. 4b), poorly temperature dependent. A value of about 125 ps is measured at 7.5 K, T_2 decreases to about 100 ps at 20 K.

The effect of the hydrogen bond on the coherence of the D$-$Cl stretching mode clearly consists in a shortening of the dephasing time. The study of dimers embedded in nitrogen matrices led to the same conclusion [10], but dephasing times associated to the dimers in argon matrices are much longer than in nitrogen matrices: T_2 is in the hundreds of ps in Ar, and in the tens of ps in N$_2$.

4 Discussion and concluding remarks

The FEL CLIO offers a good compromise between spectral and temporal resolution to study vibrational dynamics of the DCl molecule and its complexes. Nevertheless, the DFWM signals are complicated. An effort was made in order to extract all the information contained in these signals. The shape of the DFWM signals recorded for all the discussed D$-$Cl stretching modes reveals that T_2 time is equal to the pure dephasing time T_2^*. The main significant results obtained on the dephasing time involved in different interactions are summarized in Table 2.

The study of DCl isolated in nitrogen matrices revealed the effect of the lattice on the coherence time [12]. The dephasing time of monomer species showed a clear temperature dependence, highlighting a specific interaction

Table 2. Most significant dephasing times of the DCl stretching mode in different environments

x	n	Temp. (K)	$(DCl)_x$ /N_2	$(DCl)_x-(N_2)_n$ /Ar
1	1	5−7	175±30 ps	1.3±0.6 ns
	2			0.3±0.2 ns
	1	20	8±1 ps	0.15±0.05 ns
2, ν_1	1	5−7	55±5 ps	150±15 ps
		20	21±2 ps	
2, ν_2	0, 1	5−7	20±3 ps	125±20 ps
	1	20	11±2 ps	100±20 ps

between the excited stretching vibrational mode and a low frequency local mode corresponding to a coupling of the translation and the libration of the DCl molecule trapped in the nitrogen lattice. In addition, the results in solid nitrogen demonstrated, in comparison with CO molecules embedded in nitrogen matrix, that the short measured T_2 time results from the strong coupling between the DCl species and the lattice [12].

This study of complexes and clusters embedded in van der Waals solids demonstrated the effects of the specific interactions between the oscillator and its nearest neighbours. The observed dephasing time for $DCl-N_2$ complexes in argon matrix is clearly different compared to the value obtained for isolated DCl molecules trapped in solid nitrogen. The temperature evolution of T_2 time of $DCl-N_2$ complexes is compatible with dephasing processes controlled by phonons with energy in the range of tens of wavenumbers. As these dephasing times are very long, it can be considered that the van der Waals interaction between DCl molecule and nitrogen decouples the D−Cl stretching mode from the lattice influence.

With the study of mixed N_2/Ar matrices, it is possible to access to the evolution of coherence times from the one-to-one $DCl-N_2$ complex to DCl isolated in the nitrogen lattice. The first step has been obtained, going from $DCl-N_2$ to $DCl-(N_2)_2$. A size effect is clearly observed: an increase of the cluster size enhances the rate of dephasing processes, probably because several low frequency vibrational modes are coupled to the probed oscillator. Further investigations on $DCl-(N_2)_n$ clusters should provide more precise information.

DCl dimers induce a perturbation of the D−Cl stretch stronger than $DCl-N_2$ complexes because of the weak hydrogen bond between the two D−Cl molecules. This perturbation is mainly applied to the mode ν_2. More experiments have been performed in nitrogen matrices than in argon matrices. In solid nitrogen, the results lead to the following conclusions [10]:

- the dephasing times of both D−Cl modes are not governed by the coupling with the local phonon modes responsible for the dephasing process of monomers: as in the case of $DCl-N_2$ complexes in argon matrices, the specific interaction strongly modifies the coupling between the oscillator and the lattice;
- the dephasing time of the mode ν_2, corresponding to the "bonded" hydrogen stretch, is faster than that of the mode ν_1;
- T_2 time temperature dependence of the mode ν_1 is slightly more pronounced than that of the mode ν_2.

The last two features reflect a clear effect of the hydrogen bond on the dephasing time. $(DCl)_2$ is a model system to observe this effect. The only difference between the two high frequency stretching modes is the magnitude of the perturbation by the hydrogen bond. The enhancement of dephasing processes due to the hydrogen bond is also clearly observed in argon matrices, within $(DCl)_2-N_2$ species. However, the behaviour of the dimer species in solid nitrogen is different of that observed in solid argon. Much longer coherence times have been obtained in solid argon than in solid nitrogen. This solid effect could be a consequence of the different dimer geometry in argon and in nitrogen matrices [21,24]. The dimer has a planar geometry in solid argon and not in solid nitrogen. In consequence, the ν_2 to ν_1 band intensity ratio is 9 in solid argon whereas it is 1.8 in solid nitrogen [25] and the frequency shift between the monomer and the hydrogen bonded mode is larger in solid argon (40 cm^{-1} [23]) than in solid nitrogen (29 cm^{-1} [21,25]). From these remarks, one assumes that the hydrogen bond should be the strongest in dimer embedded in argon matrices. This is puzzling, if considering the effect of the strength of hydrogen bond on vibrational dephasing process. We conclude once more that the nitrogen lattice is far from an inert medium, its effect on vibrational dynamics is non negligible, even on oscillators included in hydrogen bonded clusters.

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