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Phonons in orientationally disordered neopentane $C(CD_3)_4$

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Abstract. — The phonons of deuteriated neopentane $C(CD_3)_4$ single crystals in the orientationally disordered phase were measured at $T = 173$ K by coherent inelastic neutron scattering, yielding very broad bands that spread, at a given energy, over a large portion of the Brillouin zone while sitting on an intense background. No librational mode was detected. Selection rule violations, presumably linked to disorder, were observed. The elastic constants are discussed in terms of rotational-translational coupling, and inelastic scattering results are compared with the molecular center of mass translational disorder as obtained from diffraction experiments providing a confirmation of a previous interpretation of these experiments.

1. Introduction.

Neopentane (2,2-dimethyl propane, $C(CH_3)_4$) is a well known example of an orientationally disordered (plastic) crystal. At low temperatures, it exhibits a probably ordered phase, the structure of which is still subject to discussion [1, 2]. On heating, it undergoes a transition to a highly plastic, cubic phase in which the molecules are dynamically, orientationally disordered. The transition, which is first order as observed by the heat capacity anomaly at 140 K [3] is accompanied by a high entropy change of $18.4 \text{ Jmol}^{-1}\text{K}^{-1}$. The plastic phase is stable to 256.5 K where it melts with the relatively low (for an organic compound) entropy of fusion of $12.7 \text{ Jmol}^{-1}\text{K}^{-1}$. The origin of the plastic properties lies in the compact, symmetrical molecule (Fig.1). The almost spherical shape of the molecule and the nearly uniform distribution of the 12 hydrogen atoms over its surface make it clear why the barriers to rotational motion are so easily overcome. X-ray structural studies [1] show the plastic phase to be a disordered face-centred-cubic structure (symmetry $O_h^5 - Fm\bar{3}m$; $a = 8.78 \text{ \AA}$ at 223 K). Neutron elastic

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Fig.1. — The neopentane molecule. The carbon atoms are represented in gray, the hydrogen atoms in white.

diffraction studies of a deuterated sample (lattice constant $a = 8.66 \text{ \AA}$ at 173 K) show that a molecule has 6 preferred orientations of symmetry D_{2d} [4], in accordance with previous results obtained by incoherent neutron scattering [5].

The strong intramolecular forces in the solid give rise to high frequency modes (6-90 THz) which have been studied by Raman and infrared spectroscopy [6, 7] and neutron spectroscopy using the "small- κ " method [8]. The intermolecular modes occur in the 0-5 THz frequency range. These have been investigated by neutron spectroscopy using protonated neopentane [5, 9-11]. A number of models were used to interpret these results: translational motions of the whole molecule, rotational modes about a fixed position, rotational jumps between different orientations, and rotational diffusion. A detailed discussion of the individual molecule rotational motion from a molecular dynamics simulation is given in [12]. However, because of the large incoherent scattering contribution from the hydrogen atoms, a more detailed examination of coherent neutron scattering from a deuterated single crystal specimen was necessary to examine collective excitations.

2. Single crystal preparation.

Neopentane-d₁₂ was supplied by Merck, Sharp and Dohme (purity 0.98). Vapour growth at $\sim 220 \text{ K}$ proved most suitable since crystals of reasonable size (1 to 2 cm³) could be prepared, enclosed in a silica tube. A detailed description of the growth technique of these crystals is given in [13].

Of several crystals grown, most measurements were performed on cylindrical samples of length 10 to 15 mm and diameter 10 mm. The crystals were transported from the laboratory to the reactor in an aceton-dry ice mixture, then transferred into a cryostat preset and stabilized at 173 K to avoid the melting of the material. During the opening of the cold cryostat, He gas was flushed in to prevent frost inside the cooled compartment. The cryostat allowed to maintain a temperature gradient through the sample to prevent transportation by vapour of the material from the crystal to other parts of the cell, and the subsequent destruction of the single crystal.

Because of the isotropy of the crystals, their orientation and monocrystallinity could not be checked by optical methods before the experiment. So every experiment began by trying

Fig.2. — The $(0\bar{1}1)$ plane of the neopentane reciprocal lattice together with the Brillouin zone boundaries. The dashed lines show the directions in which phonons were recorded.

to find a suitable diffusion plane inside the available angles of the goniometer heads (-20° to $+20^\circ$) and to verify that the samples were really single crystals; if they were composed of two crystals (which happened sometimes), the biggest was selected, on the condition that the orientations of the two crystals were different enough to avoid contamination of the modes of the selected crystal by modes of the other.

The specimens had mosaic spreads of full width half maximum from 0.4° to 1.3° . In most cases, a $(0\bar{1}1)$ scattering plane was used, allowing measurements in the $[001]$, $[110]$ and $[111]$ directions. A few experiments were performed in the $(0\bar{1}0)$ and $(1\bar{1}2)$ planes. Figure 2 shows the $(0\bar{1}1)$ plane of the neopentane reciprocal lattice together with the Brillouin zone boundaries and the directions in which phonons were measured. All experiments were performed at $T = 173$ K, 33 K above the order-disorder transition.

3. The triple axis experiments.

The triple axis spectrometer allows the collection of frequency scans at fixed specified values of the wavevector $\mathbf{Q}$, or of Q -scans at fixed energies, along high symmetry directions in the reciprocal lattice of the crystal. Two spectrometers of the Orphée reactor at Laboratoire Léon Brillouin were used in the experiments: 2T located on the thermal source, and 4F2 located on one of the cold sources. The neutron incident energy was selected by the (002) planes of one for 2T and two for 4F2 pyrolytic graphite monochromator crystals of mosaic spread 0.4° . The incident and scattered neutron directions were defined by Soller collimators of divergence 0.3° to 0.8° depending on the spectrometer. The instruments were used exclusively in the W configuration. The incident wavelength was fixed at $4.05 \text{ \AA}$ for 4F2 and the final wavelength at $2.45 \text{ \AA}$ for 2T; a PG graphite filter was used on the thermal beam, and a nitrogen cooled Be filter on the cold beam. With these conditions, the resolution ellipsoid had a total frequency width of approximately 0.05 THz (4F2) or 0.2 THz (2T). Only at small wavevectors ξ were the phonon peaks sharp. They broadened rapidly with increasing ξ , sitting

Fig.3. — Translational phonon measured at constant $Q(1.5, 0.5, 0.5)$.

on a strongly sloping background. This background is believed to arise from librational and rotational motions of the molecules. For demonstration we present in figure 3 a constant Q scan at $Q(1.5, 0.5, 0.5)$; the zone boundary TA phonon shows only as a shoulder on a high and rapidly varying background. Therefore, constant Q scans were restricted to small wavevectors, where the signals were reasonably strong and peaks were narrow. These measurements on 4F2 allowed to extend the experiments closer to the zone center, due to the better resolution compared to 2T. For $E \geq 0.3$ THz, we employed the constant E mode of operation in order to arrive at a better determination of the background. Some examples of constant E scans along the line $(1+\xi, 1-\xi, 1-\xi)$ are displayed in figure 4. Figure 5 summarizes the results obtained on the two spectrometers. The consistency of the results is good despite the many differences in the ways the spectra were recorded: neutron wavelength, resolution, experimental procedure, samples, and even the model used to fit the results, as will be explained in the next paragraph. Particular difficulties were encountered in the (110) direction: we were not able to identify phonon peaks beyond $\xi \simeq 0.35$. The reason for this is not clear.

4. Data analysis.

Since the data were recorded on two different spectrometers with very different experimental conditions, different analysis procedures had to be used.

Fig.4. — Transverse phonons in the [111] direction measured at constant energy. Symbols: (x) 0.48 THz, (●) 0.58 THz, (○) 0.68 THz, (Δ) 0.77 THz, (+) 0.87 THz. The lines are only guides for the eye.

4.1 DATA RECORDED ON THE COLD NEUTRON SPECTROMETER 4F2. — These were fitted by the sum of three functions:

- a δ function at $\Delta\nu = 0$ THz;
- a Lorentzian function centered at $\Delta\nu = 0$ THz to take into account the quasi-elastic scattering; and
- a symmetric damped oscillator centered at the frequency of the phonon.

These functions were convoluted with the resolution function of the spectrometer.

In some cases, as shown in figure 6, a typical spectrum in a longitudinal configuration, in addition to the expected LA phonon, another peak is seen at smaller energy. Changing the vertical collimation had no perceptible effect besides modifying the elastic contamination. This happened for longitudinal phonons in every main direction. A second damped oscillator was then added to the data analysis.

4.2 DATA RECORDED ON THE THERMAL NEUTRON SPECTROMETER 2T. — Figure 7 shows a LA($\xi\xi\xi$) phonon measured at $\nu = 0.387$ THz in the vicinity of the [111] Bragg peak. The

Fig.5. — Dispersion curves of neopentane at $T = 173$ K. The symbol (o) denotes longitudinal, (+) transverse polarisations. The lines are only guides for the eye.

configuration of the spectrometer is such that the focalised region is at $(1 + \xi, 1 + \xi, 1 + \xi)$, leading in principle to a strong, sharp peak on that side, and a lower and wider one on the other, not what is observed. This is due to the rapid variation with Q of the dynamic scattering factor of neopentane (Fig. 8). Symmetric damped oscillators thus could not be used, because of the different weight of the two sides, as the dynamic scattering factor varies significantly from one side to another.

Consequently, the data were fitted by two Lorentzian functions, with equal widths, their positions symmetric with respect to the zone center, and their intensities free to adjust. These were convoluted with the resolution function of $2T$.

Despite different samples, spectrometers, neutron wave lengths, experimental procedures and data analysis methods, the results are consistent, as mentioned above, not only for the frequencies of the phonons (cf. Fig. 5), but also to a degree for their widths (HWHM) in energy (cf. Fig. 9).

5. Discussion.

5.1 SELECTION RULE VIOLATIONS. — At small wavevectors, scans performed in purely longitudinal configuration exhibited besides the expected LA phonon an additional peak of strong intensity at lower frequency (see Fig.6). As the extra peak occurred in all symmetry directions under various experimental conditions, we can rule out experimental artifacts. We believe it is due to contamination from transverse phonons, although it is not at exactly the same frequency, caused by translational disorder as their strong intensities cannot be accounted for by vertical divergence of the beam. It should be pointed out that in this crystal, the molecular center of mass translational disorder is quite large, even at a relatively low temperature: when the translational disorder $\langle u_z^2 \rangle_{\text{phonon}}$ due to phonons is expected to be less than $0.05 \text{ \AA}^2$ it was

Fig.6. — Longitudinal phonon in the [001] direction. The phonon, its symmetric are pointed at by arrows. The line is the result of a least square fit as described in the text. The additional mode is interpreted as a violation of selection rules.

found that $\langle u_x^2 \rangle \simeq 0.1 \text{ \AA}^2$ [4]. This was understood as resulting from the extra volume required by reorienting molecules. The contamination may thus result from a selection rule violation, caused by local configuration fluctuations, the fcc lattice being only an average.

5.2 ROTATIONAL-TRANSLATIONAL COUPLING. — We now discuss the elastic constants in terms of the rotational-translational (RT) coupling and compare the inelastic scattering data with previous diffraction results.

The elastic constants were obtained from the data shown in table I. A least square fit of the three elastic constants C_{11} , C_{12} , and C_{44} to the six measured sound velocities was done, using the usual relations in a cubic crystal and the volumic mass of deuterated neopentane $\rho = 859.4 \text{ kg m}^{-3}$. The differences between measured sound velocities and those calculated from elastic constants are well within error bars.

The elastic constants of neopentane obtained in this manner are given in table II along with those of some other plastic crystals. We also give the acoustic anisotropy: $A = 2C_{44}/(C_{11} - C_{12})$, the deviation from the Cauchy rule: $\delta = (C_{44} - C_{12})/C_{12}$, which is a rough measure of the RT coupling, and, for tetrahedral molecules, the symmetry of the equilibrium orientations and the molecular center of mass translational disorder from diffraction results.

The anisotropies of noble gasses Ne, Ar, Kr, Xe, for which there is clearly no orientational disorder, are in the range $2.4 \sim 2.7$ and $|\delta| \simeq 0.05 \sim 0.28$ (see [14]). The situation seems clear for adamantane, as both A and δ are close to noble gas values and the RT coupling as obtained from diffraction [15] is negligible. The T_d molecules sitting with T_d orientations⁽¹⁾ on O_h sites, behave, if this line of thinking is correct, basically as spheres.

⁽¹⁾ i.e. with the three $\bar{4}$ molecular symmetry axes parallel to the crystal 4-fold axes.

Fig.7. — The symbol (o) denotes a longitudinal phonon in the [111] direction, its symmetric as a function of reduced wave vector. The continuous line is given by the resolution function, convoluted with a symmetric mode.

Fig.8. — Dynamic scattering factor as a function of reduced wave vector.

On the other hand, for CD_4 , the value of δ is large enough not to be neglected, and indeed an interpretation of the phonon properties, on the basis of RT coupling, is given in references [14, 17]. It must be pointed out, however, that the orientational probability density function $P(\Omega)$ as obtained from diffraction experiments is essentially isotropic, with no hint of RT coupling.

Finally, the three compounds with D_{2d} equilibrium orientations⁽²⁾, neopentane, CCl_4 , and

⁽²⁾ i.e. only one $\bar{4}$ molecular axis is parallel to one of the crystal axes, the other two being parallel to crystal 2-fold axes.

Fig.9. — Phonon linewidths with the same conventions as for the dispersion curves. The lines are only guides for the eye.

Table I. — The first line shows the measured frequency ν_{obs} , with uncertainties (second line), in each direction, for a value of q (third line) chosen so that the approximation that the corresponding branch is linear, is reasonable, and uncertainties (fourth line). The sound velocities v_{obs} calculated with the lattice constant $a = 8.66 \pm 0.20 \text{ \AA}$ are on the fifth line. The last two lines give v_{calc} and ν_{calc} obtained from the elastic constants, showing the consistency of this procedure. The sound velocity for the [011] T_2 branch is also calculated although it could not be measured because undistinguishable from the foot of the elastic contribution in the case when the sample orientation would have allowed such a measurement.

	[100]		[011]			[111]	
	L	T	L	T_1	T_2	L	T
ν_{obs} [THz]	1.40	0.95	1.35	0.79		1.45	0.97
$\delta\nu_{\text{obs}}$ [THz]	0.05	0.05	0.05	0.05		0.05	0.05
q	0.50	0.60	0.335	0.35		0.27	0.45
δq	0.01	0.01	0.01	0.01		0.01	0.01
v_{obs} [m/s]	2425	1371	2468	1382		2685	1078
δv_{obs} [m/s]	191	127	222	159		254	104
v_{calc} [m/s]	2330	1400	2570	1400	860	2650	1070
ν_{calc} [THz]	1.34	0.97	1.41	0.80		1.43	0.96

CB₄, have essentially the same rather large value of δ , thus pointing in the direction of much stronger for CB₄ ($\tilde{C}_{31} = 0.10$, $\tilde{C}_{71} \simeq 0.0$, $\tilde{C}_{72} = -0.13$) [23]⁽³⁾ than for neopentane ($\tilde{C}_{31} = 0.03$, $\tilde{C}_{71} \simeq 0.0$, $\tilde{C}_{72} = 0.03$) [4].

At this point, the connection between RT coupling as obtained from diffraction experiments and from inelastic scattering is not clear. Yet, one should keep in mind that a diffraction experiment yields the single molecule probability density function $P(\mathbf{u}, \Omega)$, where $\mathbf{u}$ is the molecular displacement with respect to its mean position, and Ω its orientation as given for

⁽³⁾ These coefficients come from the expression of $P(\mathbf{u}, \Omega)$ as: $P(\mathbf{u}) (P(\Omega) + P^{[1]}(\Omega) \cdot \mathbf{u} + \dots)$ and the expansion of the first order coupling term $P^{[1]}(\Omega)$ on the correct set of symmetry adapted rotator functions [24].

Table II. — *Elastic constants of neopentane (GPa) along with those of some other plastic crystals. The last four columns give the acoustic anisotropy A , the deviation from the Cauchy rule δ , for tetrahedral molecules the symmetry of the equilibrium orientations and the translational disorder ($\text{\AA}^2$).*

	C_{11}	C_{12}	C_{44}	A	δ	Orient.	$\langle u_x^2 \rangle$
C(CD ₃) ₄ (this work)	4.65	3.37	1.68	2.63	-0.50	D _{2d}	0.1
Adamantane [15, 16]	7.59	4.55	3.44	2.26	-0.24	T _d	0.046
CD ₄ (I) [18, 19]	2.056	1.542	0.939	3.65	-0.39	Isotr.	0.067
CCl ₄ (Ia) [20, 21]	3.569	3.163	1.111	5.47	-0.65	D _{2d}	$\simeq 0.3$
CBr ₄ (Brillouin) [22]	4.221	3.357	1.613	3.73	-0.52	D _{2d}	
CBr ₄ (neutrons) [23]	7.68	3.88	2.08	1.09	-0.46	D _{2d}	0.19
SF ₆ [29]	3.409	2.218	1.323	2.22	-0.40		
Succinonitrile [27]	5.07	3.54	0.65	0.85	-0.82		
KCN [28]	19.16	11.97	1.405	0.39	-0.88		
NaCN [30]	25.34	14.44	0.33	0.06	-0.98		

instance by Euler angles. $P(\mathbf{u}, \Omega)$ can be converted into the potential $V(\mathbf{u}, \Omega)$ of a molecule in the average field created by its neighbours. On the other hand, an inelastic scattering experiment is mostly sensitive to the two molecule interaction potential $V(\mathbf{u}_1, \Omega_1, \mathbf{u}_2, \Omega_2)$, which in [14, 17] is averaged to $V(\mathbf{u}_1, \Omega_1, \mathbf{u}_2)$ and summed over all first neighbours of molecule 1.

This can be related to the fact that diffraction experiments done on neopentane yield a large value of the molecular center of mass translational disorder $\langle u_x^2 \rangle$ which was interpreted as the effect of a small proportion of molecules in the process of reorientation pushing aside their neighbours [4, 25]. This factor was, in neopentane, by no means negligible, indeed it was the dominant feature of the diffraction results. If it were to be understood in terms of an interaction potential, this would be the coupling of the orientation of molecule 1 with the displacement of molecule 2, $V(\Omega_1, \mathbf{u}_2)$.

Thus, in order to compare RT coupling as obtained from inelastic scattering experiments with diffraction results, it is $\langle u_x^2 \rangle$, not the RT terms in the expansion of $P(\mathbf{u}, \Omega)$, that is the correct quantity. This is confirmed in the last column of table II in which there is a remarkably good correlation of $\langle u_x^2 \rangle$ with $|\delta|$. Adamantane has small values of $|\delta|$ and $\langle u_x^2 \rangle$, CD₄ slightly larger, neopentane and CBr₄ intermediate and CCl₄ large.

This confirms, from a completely independent point of view our previous interpretation of diffraction results obtained on neopentane. It would be interesting at this point to compare these results with diffuse scattering experiments in which one obtains in principle $P(\mathbf{u}_1, \Omega_1, \mathbf{u}_2, \Omega_2)$.

This rather qualitative discussion needs however to be backed with calculations in the line of [14, 17] or [26], which are beyond the scope of the present paper. This would have the additional advantage of including the bandwidths in the discussion.

6. Conclusion.

The phonons of deuteriated neopentane in the orientationally disordered phase were measured on several different samples on two different spectrometers with different recording techniques. Careful analysis of the data provide consistent results, despite the difficulty to characterize some of the phonons that spread, at a given energy, over a large portion of the Brillouin zone

while sitting on an intense background.

While no librational mode could be detected, selection rule violations for small wave vectors, presumably linked to disorder, were observed.

A qualitative discussion allows to understand the elastic constants in terms of the coupling of the orientation of molecule 1 with the translation of molecule 2, links inelastic scattering results and the molecular center of mass translational disorder as obtained from diffraction experiments and confirms from a very different point of view the previous interpretation of these experiments.

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