

Local Order in Fully Deuterated Liquid *N*-Methylacetamide ($\text{C}_3\text{D}_7\text{NO}$) As Studied by Neutron Diffraction and Density-Functional Theory Calculations

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A structural investigation of fully deuterated liquid *N*-methylacetamide ($\text{NMA}_{\text{d}7}$) is performed at 308 K and atmospheric pressure by using neutron diffraction together with density-functional theory (DFT). The analysis of experimental data yields the total structure factor $S_{\text{M}}(Q)$, the molecular form factor $F_1(Q)$, and the distinct pair correlation function $g_{\text{L}}(r)$. The DFT calculations are performed to study the relative stability of the two possible isomers (trans and cis) and to examine some possible clusters recently published that may describe the intermolecular arrangement in the liquid state. Neutron measurements can be interpreted in terms of trans linear trimer (T1) and cis cyclic trimer (T2) where the total number of hydrogen bonds is respectively equal to two and three. The theoretical structure factors obtained on the basis of intermolecular arrangements agree fairly well with the experimental one beyond $Q = 2 \text{ \AA}^{-1}$. All through the study, a comparison is made with complementary X-ray results.

Introduction

Hydrogen bonding of peptide groups is considered to be an important factor in determining the three-dimensional structure of biological macromolecules such as polypeptides and proteins.¹ Because of the presence of a peptide group, *N*-methylacetamide (NMA) has been recognized as a useful model compound and has been extensively studied.^{2–8}

The structures of *trans*-NMA in the crystal and in the gas phase were determined by X-ray⁹ and electron^{10,11} diffraction experiments, respectively. A comparison between the bond distances of the planar molecules in both phases shows a lengthening of C–N and shortening of C=O bonds in the gas phase. The increase and decrease of the C–N and C=O bond lengths are due to hydrogen bond formation in the crystalline state. The crystal structure⁹ of NMA consists of linear chains in which two hydrogen bonds (2.825 Å in length) between CO and NH groups connect each molecule to adjacent ones. A disordered structure was observed between 238 and 301 K. At higher temperature, neat NMA is in its liquid phase. This is believed to have an even more disordered structure, through one still maintaining some degree of hydrogen bonding.⁶ However, none of the earlier studies have experimentally examined the problem of local order in liquid NMA.

So, we have performed a first structural investigation of liquid NMA by X-ray scattering at 308 K. Our published measurements clearly show that the local order in the liquid state is comparable to that existing in the crystalline state.¹² To examine in more detail the intermolecular association in liquid NMA, X-ray data were analyzed in a second paper¹³ in relation to DFT

calculation. A large variety of dimers and trimers including cis and trans monomers were analyzed to describe the structure of the liquid. In particular, it has been found that the more probable clusters that can reproduce the local arrangement in liquid NMA are a ring cis trimer and a chainlike trans trimer.

To get further insight into the local order of liquid NMA, the problem of intermolecular arrangement is studied in this paper by means of neutron diffraction in combination with DFT calculations. Technical details about this theoretical method are described in our previous study.¹³ In the latter paper, we were not interested in the hydrogen atom positions, only the X–Y ($X \neq \text{H}$, $Y \neq \text{H}$) correlations were considered; whereas in the current paper all the interatomic correlations in fully deuterated NMA are taken into account. So, we have improved the basis set by adding a polarization function of type p for H atoms. To our knowledge, no experimental information related to local order in fully deuterated liquid *N*-methylacetamide ($\text{NMA}_{\text{d}7}$) occurs in the literature and this is the first attempt to apply neutron scattering formalism to the study of this problem. Experimental data are first analyzed to yield the total structure factor and the intermolecular pair correlation function. Second, the theoretical intermolecular distances for the two possible isomers are considered in order to construct the molecular form factors. Third, the substitution effect H/D on the intra- and intermolecular interactions is examined. Fourth, the more probable models as deduced from our previous X-ray study¹³ are reexamined to describe the intermolecular arrangement in deuterated liquid NMA. The computed structure factors and pair correlation functions of these models are then compared with those extracted from neutron scattering data.

Experimental Details

The fully deuterated *N*-methylacetamide ($\text{NMA}_{\text{d}7}$) was supplied by C/D/N isotopes (chemical purity 99+%). In the experiments, $\text{NMA}_{\text{d}7}$ was used without further purification. The

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sample holders were filled and sealed in a glovebox to avoid contamination by atmospheric water.

The neutron scattering experiments were performed at the reactor Orphée of the Laboratoire Léon Brillouin, at Saclay (France), using the 7C2 spectrometer. All details about this spectrometer are reported by Ambroise, Bellissent, and Bellissent-Funel.^{14,15} This spectrometer is equipped with a BF_3 position sensitive detector with 640 cells; the angular step between two adjacent cells is equal to 0.2° , which leads to a maximum diffraction angle of 128.7° . We select an incident wavelength of $\lambda = 0.711 \text{ \AA}$ by means of a Cu (111) monochromator. The sample is contained inside a null matrix $\text{Ti}_{67}\text{-Zr}_{33}$ cell for which the intensity is principally incoherent. The cell has an internal diameter $\phi_{\text{int}} = 5.70 \text{ mm}$ and a wall thickness $e = 5.22 \text{ mm}$. All experiments are carried out at 308 K. The usual measurements of the vanadium rod ($I_{\text{v}}^{\text{expt}}$), the cadmium rod ($I_{\text{cd}}^{\text{expt}}$), and the empty cryostat ($I_{\text{vacuum}}^{\text{expt}}$) were also performed.

Results and Discussions

The Experimental Structure Factor. The theoretical framework for neutron scattering by molecular liquids is discussed in previous studies.^{16–20} All necessary details about the data treatment and the fitting procedures permitting extraction of respectively the total structure factor $S_{\text{M}}(Q)$, the molecular form factor $F_{\text{I}}(Q)$, and the H-bonding function $D_{\text{M}}^{\text{HB}}(Q)$ to model the intermolecular arrangement in the liquid can be found in refs 21 and 22.

The measured intensities for the cell filled with deuterated liquid NMA, the empty cell, the empty cryostat, the vanadium rod, and the cadmium rod are represented in Figure 1A, A1–A5. Figure 1B (solid line) shows the differential scattering cross section ($d\sigma/d\Omega$) of the liquid after the analytic corrections and vanadium normalization. The incoherent scattering term was also subtracted. The dashed line gives the inelasticity correction $P(Q)$, computed according to a polynomial procedure described elsewhere.^{21,22} The total structure factor of liquid NMA_{d7} is drawn in Figure 2, parts A and B (solid line). The dashed line in Figure 2A,B gives the contribution $S_{\text{M}}(\infty)$ of the independent molecules.¹²

Intramolecular Structure. As mentioned above, we have considered the two possible trans and cis monomers. The molecular form factors $F_{\text{I}}(Q)$ [Figure 2, parts A and B, respectively, dotted line] for these isomers are calculated from analytical expressions after the fitting procedure described elsewhere.^{13,21,22} One can see a good agreement between the experimental structure factor $S_{\text{M}}(Q)$ [Figure 2A,B, solid line] and the computed molecular form factors at high Q -range. In our fitting procedure and to reduce the number of adjustable parameters, the intramolecular distances are kept equal to the deduced values from our DFT geometry optimization; concerning the Debye–Waller (DW) parameters, the coefficients recently deduced¹³ by analyzing our X-ray scattering data on liquid NMA at 308 K are also retained. However, it is well-known that in the case of X-ray scattering, the contributions of X–H correlations (with $\text{X} = \text{H}, \text{C}, \text{N}$, and O) to the molecular form factor are very low and sometimes neglected at high Q -range.²³ This hypothesis is never valid in the case of neutron scattering. The hydrogen atoms are substituted by the deuterium ones which have a similar coherent scattering section to carbon, oxygen, or sodium. Then, all the DW parameters are considered here.

The experimental pair correlation function $g(r)$ as deduced from neutron scattering is plotted in Figure 3A,B (solid line)

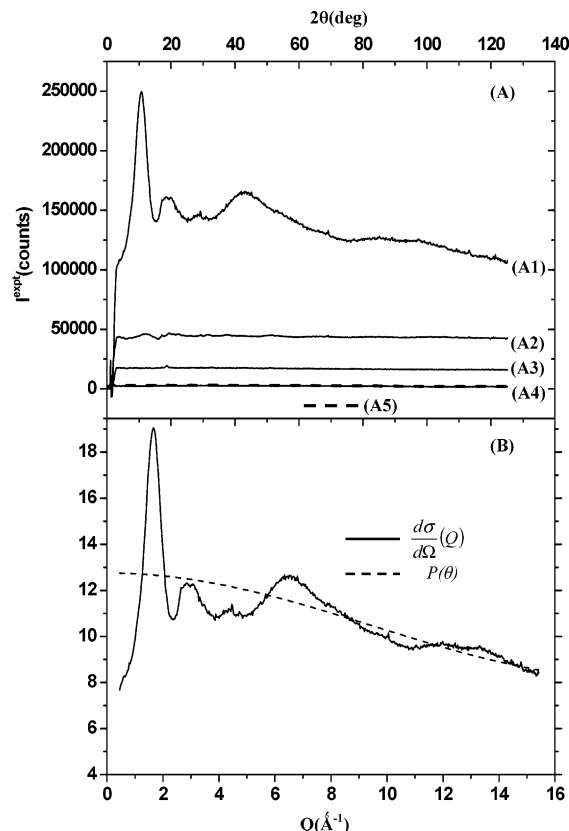


Figure 1. (A) Measured intensities $I^{\text{expt}}(2\theta)$ for (A1) a cell filled with deuterated liquid *N*-methacetamide, (A2) the empty cell, (A3) the vanadium rod, (A4) the empty cryostat, and (A5) the cadmium rod (dashed line). (B) Differential neutron scattering cross section for liquid NMA_{d7} . The dashed line is the inelastic correction $P(Q)$ calculated by using a polynomial expansion.

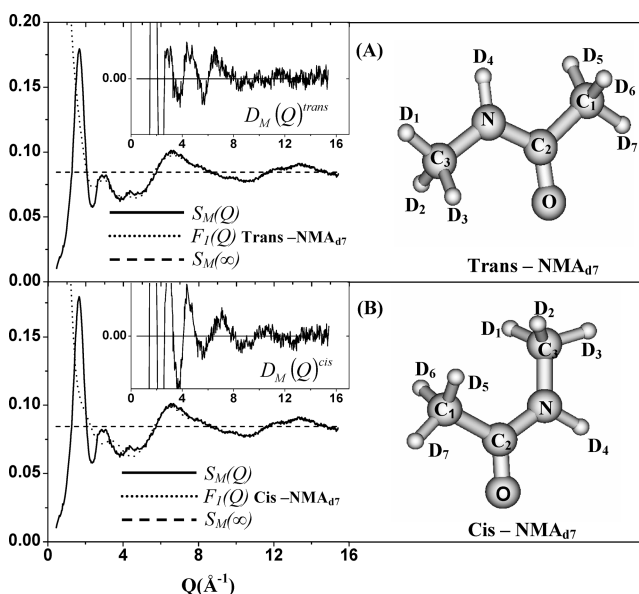


Figure 2. The total structure factor $S_{\text{M}}(Q)$ (solid line) of deuterated liquid *N*-methacetamide, derived from neutron scattering at 308 K with the molecular form factors $F_{\text{I}}(Q)$ (dotted line) of trans and cis isomers (A and B, respectively). The dashed line gives the asymptotic value $S_{\text{M}}(\infty)$ of the total structure factor at large Q range. Inset: The intermolecular structure functions $D_{\text{M}}(Q)$. The oscillations seen at $(5\text{--}9 \text{ \AA}^{-1})$ Q -range are due to the H-bonding correlations.

together with the Fourier transformations of the molecular form factors of the trans and cis isomers, i.e., $g_{\text{intra}}(r)^{\text{trans}}$ and $g_{\text{intra}}(r)^{\text{cis}}$ [Figure 3A,B, dotted line]. Similar curves as deduced from

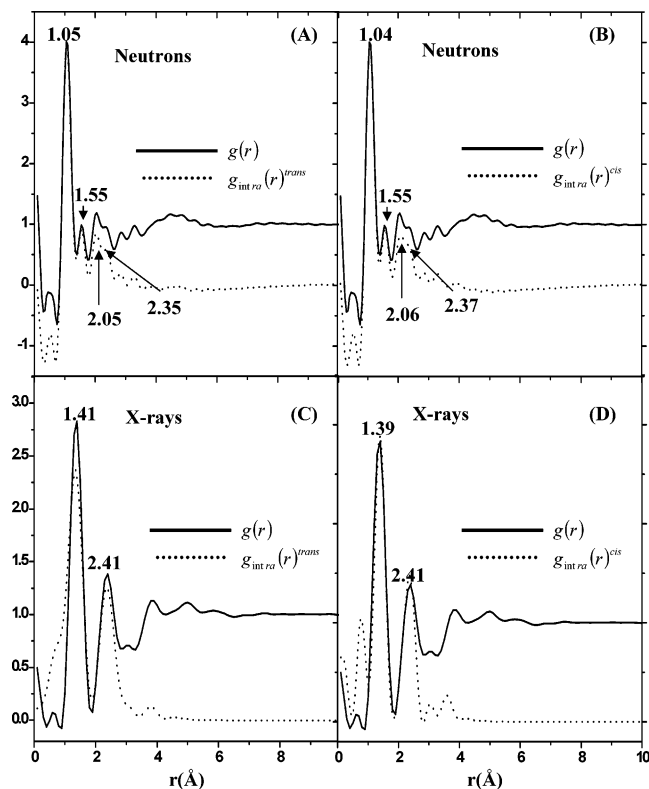


Figure 3. The neutron and X-ray pair correlation functions $g(r)$ (solid line) of liquid NMA are compared to the intramolecular pair correlation curves $g_{\text{intra}}(r)^{\text{trans}}$ and $g_{\text{intra}}(r)^{\text{cis}}$ (dotted line) of trans and cis conformers (A, C and B, D, respectively). The indicated r_{ij} values correspond to dotted curves.

our previous X-ray study¹³ are also drawn in Figure 3C,D for comparison. One can notice that the theoretical curves reproduce the experimental ones well at low r -range. It is also interesting to underline the following features of the curves. First, neutron results show, in the case of trans monomer, the appearance of two peaks at respectively 1.05 and 2.05 Å which are nonexistent in the X-ray case. The first peak is attributed to some intramolecular C–D ($\text{C}_3\text{--D}_1 = 1.089$ Å, $\text{C}_3\text{--D}_3 = 1.093$ Å, ...) and $\text{N}\cdots\text{D}$ ($\text{N}\cdots\text{D}_4 = 1.005$ Å) bonds, whereas the second one is ascribed to other $\text{N}\cdots\text{D}$ correlations ($\text{N}\cdots\text{D}_1 = 2.084$ Å, $\text{N}\cdots\text{D}_2 = 2.095$ Å, ...) and $\text{C}_1\cdots\text{D}_1 = 2.048$ Å nonbonding interactions within a NMA molecule. Second, the observed peaks at 1.41 and 2.41 Å in the X-ray $g_{\text{intra}}(r)^{\text{trans}}$ curve are respectively shifted to 1.55 and 2.35 Å in the neutron $g_{\text{intra}}(r)^{\text{trans}}$ curve. This is due to some D–D ($\text{D}_5\text{--D}_6 = 1.768$ Å, $\text{D}_5\text{--D}_7 = 1.777$ Å, ...) and $\text{D}_4\cdots\text{D}_7 = 2.326$ Å contributions. A similar trend may be outlined about the cis isomer curves.

Intermolecular Structure. The intermolecular structure factors $D_{\text{M}}(Q)^{\text{trans}}$ and $D_{\text{M}}(Q)^{\text{cis}}$ deduced by subtracting the molecular form factors $F_1(Q)^{\text{trans}}$ and $F_1(Q)^{\text{cis}}$ from the total structure factor $S_{\text{M}}(Q)$ are plotted in the inset in Figure 2A,B (solid line), respectively. The observed oscillations approximately in the 5–10 Å^{−1} Q -range are the signature of hydrogen bonds in liquid *N*-methylacetamide. It is well-known that in the case of simple liquids such as DMF,²⁴ CCL₄, and CS₂,^{25,26} the function $D_{\text{M}}(Q)$ decays to zero very rapidly. Almost no oscillations could be detected for the large Q -range. On the basis of this argument, we assume that at high Q values, the $\text{N}\cdots\text{O}$ and $\text{D}\cdots\text{O}$ H-bond interactions contribute most to the $D_{\text{M}}(Q)$ functions, i.e.

$$D_{\text{M}}(Q) \cong D_{\text{M}}^{\text{HB}}(Q) = D_{\text{N}\cdots\text{O}}^{\text{HB}}(Q) + D_{\text{D}\cdots\text{O}}^{\text{HB}}(Q) = 2n \left\{ b_{\text{N}} b_{\text{O}} \frac{\sin(Qr_{\text{NO}})}{Qr_{\text{NO}}} \exp\left[-\frac{\mu_{\text{NO}}^2 Q^2}{2}\right] + b_{\text{D}} b_{\text{O}} \frac{\sin(Qr_{\text{DO}})}{Qr_{\text{DO}}} \exp\left[-\frac{\mu_{\text{DO}}^2 Q^2}{2}\right] \right\} / \left(\sum_{\alpha=1}^m b_{\alpha}^2 \right) \quad (1)$$

where $\mu_{\alpha\beta}$ is the root-mean-square (rms) vibrational amplitude or the DW parameters of the site–site separated distance and n is the total number of hydrogen bonds in the considered intermolecular aggregates.

By a least-squares fitting of expression 1 to the large Q part ($Q > 5$ Å^{−1}) of $D_{\text{M}}(Q)^{\text{trans}}$ and $D_{\text{M}}(Q)^{\text{cis}}$, the structural parameters of the H-bond interactions are deduced. In this calculation, the total number n of H-bonds is assumed to be the same as that recently found¹³ in the more probable clusters in the NMA liquid, i.e., $n = 2$ and 3. In fact, among a large variety of models including dimers and trimers, we have shown by combining X-ray data and DFT calculation that the local order in liquid NMA is particularly reproduced by considering a chainlike trans trimer or a ring cis trimer. Figure 4A shows respectively the Fourier transformations $g_{\text{L}}^{\text{HB}}(r)^{\text{trans}}$ and $g_{\text{L}}^{\text{HB}}(r)^{\text{cis}}$ of the $D_{\text{M}}^{\text{HB}}(Q)^{\text{trans}}$ and $D_{\text{M}}^{\text{HB}}(Q)^{\text{cis}}$ functions as deduced by considering, respectively, the trans (solid line) and cis (dotted line) isomers. Similar curves based on X-ray scattering data¹³ are also reported in Figure 4B. It is important to underline that in our fitting procedure of neutron data, the $\text{N}\cdots\text{O}$ and $\text{D}\cdots\text{O}$ distances are deduced from DFT calculation, the DW parameters $\mu_{\text{N}\cdots\text{O}}$ are considered from our previous X-ray study,¹³ and only the $\mu_{\text{D}\cdots\text{O}}$ ones are fitted here. The hydrogen bond parameters are quoted in Table 1. By comparing parts A and B of Figure 4, we can clearly notice the contribution of the $\text{X}\cdots\text{D}$ interaction in the $D_{\text{M}}^{\text{HB}}(Q)$ function. The observed peaks respectively at 2.95 and 2.85 Å in $g_{\text{L}}^{\text{HB}}(r)^{\text{trans}}$ and $g_{\text{L}}^{\text{HB}}(r)^{\text{cis}}$, assigned to the $\text{N}\cdots\text{O}$ interactions, are more manifest in the X-ray curves,¹³ whereas neutron results show two more pronounced peaks at 1.95 Å, corresponding to $\text{D}\cdots\text{O}$ interactions. It is interesting to underline, here, the complementarity between the information that can be extracted from X-ray and neutron scattering.

In Figure 4C,E, we compare respectively the Fourier transformations of $D_{\text{M}}(Q)^{\text{trans}}$ and $D_{\text{M}}(Q)^{\text{cis}}$, i.e., $g_{\text{L}}(r)^{\text{trans}}$ and $g_{\text{L}}(r)^{\text{cis}}$ (solid line) as deduced from neutron data to the computed $[g_{\text{L}}^{\text{HB}}(r) - 1]$ values (dotted line) of $D_{\text{M}}^{\text{HB}}(Q)^{\text{trans}}$ and $D_{\text{M}}^{\text{HB}}(Q)^{\text{cis}}$. Similar curves corresponding to X-ray investigations¹³ are reported in Figure 4D,F. Two important points should be addressed. On one hand, the complementarity between X-ray and neutron experiments information is evident. In fact, the $\text{N}\cdots\text{O}$ and $\text{D}\cdots\text{O}$ signature of hydrogen bonding in liquid NMA is respectively better described via the former and the latter techniques. On the other hand, the H-bonding parameters as optimized from DFT calculation agree fairly with experimental results.

The residual discrepancy between the experimental structure factor $S_{\text{M}}(Q)$ and the molecular form factors, associated with H-bonding contribution, i.e., $[F_1(Q) + D_{\text{M}}^{\text{HB}}(Q)]$ functions [see the oscillations in the $D_{\text{M}}^{\text{NHB}}(Q)$ functions, inset in Figure 5A,B (solid line)] are ascribed to intermolecular correlations other than those of H-bonding. To describe these oscillations, we have considered the two more probable molecular arrangements described above: the chainlike trans trimer (T_1) and the cyclic cis trimer (T_2) [Figure 5, parts A and B, respectively].

In parts A and B of Figure 5, we compare the experimental structure factor $S_{\text{M}}(Q)$ (solid line) of liquid deuterated NMA to

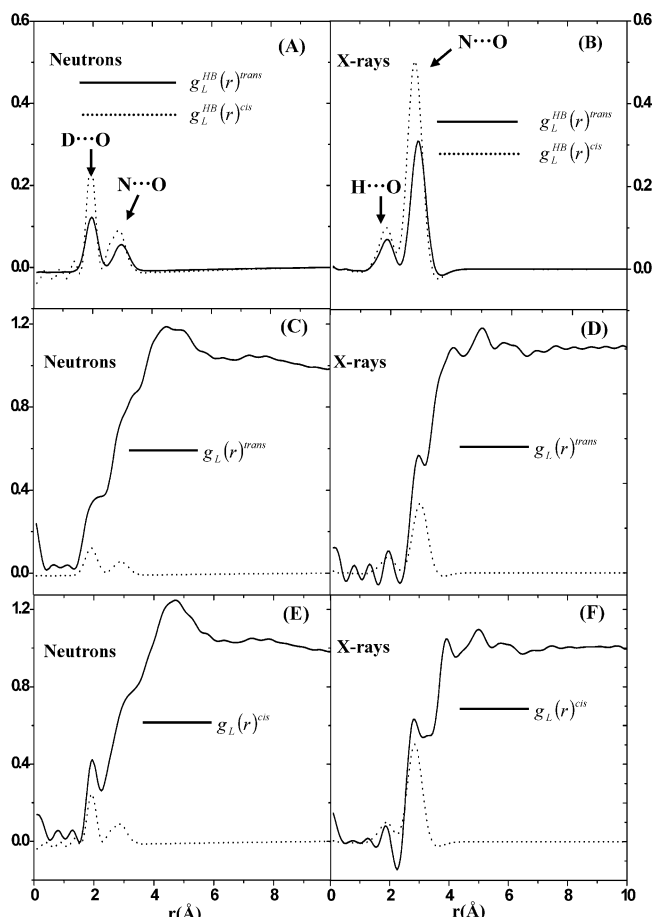


Figure 4. The neutron and X-ray Fourier transformations of the $D_M^{\text{HB}}(Q)$ functions as deduced by considering respectively the isomers cis (dotted line) and trans (solid line) (A and B, respectively). The $g_L(r)$ pair correlation functions derived at 308 K (solid line) from neutron scattering measurements (C, E) and X-ray data (D, F) by considering the isomers trans and cis. The dotted line shows the contribution $[g_L^{\text{HB}}(r) - 1]$ of H-bond interactions inside a chainlike trans trimer and a ring cis trimer (C, D and E, F, respectively).

TABLE 1: H-Bonding Parameters of Deuterated Liquid NMA at 308 K As Deduced from X-ray and Neutron Scattering Data and by Considering Respectively the Isomers Trans and Cis

$r^b_{\text{N}\cdots\text{O}}$	$\mu^a_{\text{N}\cdots\text{O}}$	$r^b_{\text{D}\cdots\text{O}}$	$\mu^a_{\text{D}\cdots\text{O}}$
trans			
2.967	0.249	1.971	0.197
2.877	0.249	1.973	0.197
cis			
2.859	0.249	1.941	0.100
2.857	0.249	1.934	0.101
2.857	0.249	1.939	0.347

^a X-ray scattering data. ^b DFT calculation, the total number of H-bonds is respectively equal to two and three.

the theoretical ones $S_M^{\text{C}}(Q)$ as deduced from these considered models. In these calculations, the intermolecular distances are fixed and taken from the DFT optimized geometry of each cluster; only the DW parameters are fitted. Manifestly, the proposed models describe well the local order in the liquid since the mean features of the experimental structure factor are largely reproduced by the computed ones beyond $Q = 2.5 \text{ \AA}^{-1}$. The dashed line [Figure 5A,B] represents the residual difference between $S_M(Q)$ and $S_M^{\text{C}}(Q)$ curves.

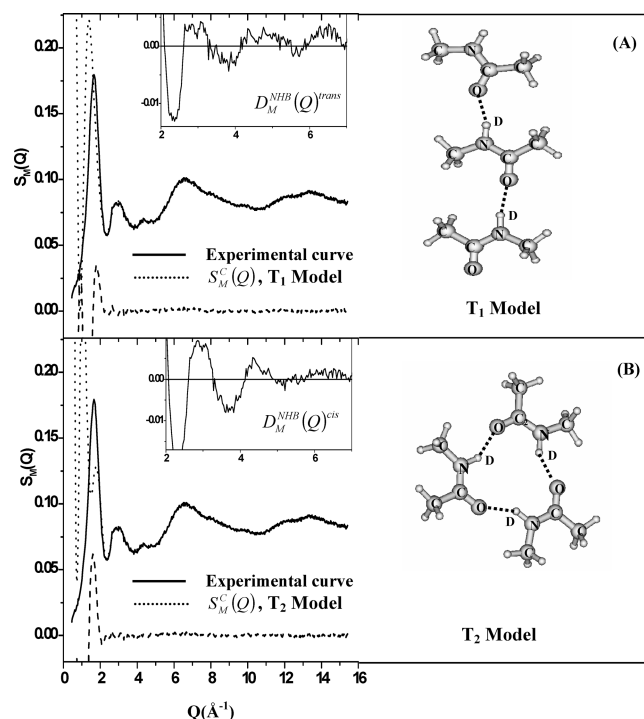


Figure 5. The experimental structure factor $S_M(Q)$ (solid line) of liquid NMA_{d7} as deduced from neutron scattering experiment, compared to the computed $S_M^{\text{C}}(Q)$ values (dotted line) relative to the trimers T₁ and T₂ (A and B, respectively). The dashed line shows the difference between the experimental and the theoretical curve. The oscillations seen at the low Q -range in the intermolecular structure functions $D_M^{\text{HB}}(Q)$ (inset) are due to intermolecular correlations other than those of H-bonding.

Conclusion

In this study, we have presented a complementary structural investigation of fully deuterated liquid *N*-methylacetamide by means of neutron scattering and DFT calculations. On one hand, neutron diffraction data are used to deduce, after corrections and normalization of the cross section, the structure factor $S_M(Q)$ and the pair correlation functions $g(r)$ of the liquid. To our knowledge, this is the first study where the local order in liquid NMA is approached via neutron scattering technique, whereas many studies related to similar liquids can be found in the literature. On the other hand, DFT results are used to construct the theoretical form factors of the two possible isomers and the total structure factors of two proposed models. It is interesting to note that in the current neutron scattering study, all the interatomic interactions are taken into account, whereas in our previous study,¹³ based on X-ray scattering data, only the X–Y ($X \neq \text{H}$, $Y \neq \text{H}$) correlations were considered. The results of our investigation can be summarized as follows:

(a) The contribution of the intramolecular interactions in liquid NMA is well accounted for by either trans or cis molecule. In fact, X-ray data,¹³ just as neutron data, are compatible with the corresponding deduced molecular form factor from these geometries.

(b) By admitting the H-bonding interactions inside the chainlike trans trimer and ring cis trimer, a good agreement between the distinct pair correlation curves $g_L(r)^{\text{trans}}$ and $g_L(r)^{\text{cis}}$, derived by Fourier transformations of $D_M(r)^{\text{trans}}$ and $D_M(r)^{\text{cis}}$, and the computed $[g_L^{\text{HB}}(r) - 1]$ values of $D_M^{\text{HB}}(Q)^{\text{trans}}$ and $D_M^{\text{HB}}(Q)^{\text{cis}}$ functions is observed. In this calculation, the hydrogen bond distances, quantities which are generally difficult to determine experimentally, are deduced via DFT calculations.

Only the DW parameters $\mu_{D\cdots O}$ are adjusted; the $\mu_{N\cdots O}$ values are taken from our recent X-ray data analysis.¹³ In fact, X-ray results have principally shown the N \cdots O interaction, characteristic of H bonding in liquid NMA, while neutron experiments clearly highlight the second signature of H bonding, i.e., the deuterium–oxygen interaction.

(c) To analyze interactions other than those of H-bonding, we have considered in this study the two more probable clusters that we have recently found by combining X-ray scattering data and DFT calculations. Theoretical structure factors deduced from either a linear trans trimer or a ring cis trimer and which particularly take into account all the D \cdots X (X = D, N, C, O) interactions are in good agreement with the experimental neutron structure factor.

(d) The complementarity between X-ray and neutron scattering experiments should be underlined. In fact, the structural investigation of liquid NMA by using the two techniques gives corroborating results and allows us to deduce some reliable information about the local order in this liquid.

The study of temperature and pressure effect on hydrogen bonding in liquid NMA by means of X-ray and neutron scattering will be addressed in our next paper.

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