

Correlation of reorientational jumps of water molecules in bulk water

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(Received 23 January 2013; revised manuscript received 18 April 2013; published 24 May 2013)

Recent theoretical and experimental studies suggested the large-amplitude angular jump mechanism of the reorientational motions of water molecules. In this paper, we study the correlation effects of such angular jump motions, which are important for understanding a number of biological processes involving the motions of water molecules, by using molecular dynamics simulations. The results show that large-angular jump motions of a water molecule can enhance the successive jump motions of the same water molecule and the surrounding water molecules. Such a correlation can extend up to a distance of two water layers and is propagated through the perturbations to the local hydrogen bond networks. A detailed molecular picture of the correlation propagation is shown based on the molecular simulations.

DOI: [10.1103/PhysRevE.87.052309](https://doi.org/10.1103/PhysRevE.87.052309)

PACS number(s): 61.20.-p, 65.20.-w, 87.90.+y, 87.15.-v

I. INTRODUCTION

Water molecules play vital roles in biological processes. In addition to the network structures of hydrogen bonds (HBs), biological processes also rely on the dynamic properties of water molecules. For example, dynamic rearrangement and reconstruction of HB networks due to reorientational motions of water molecules are crucial for some transportations [1–4] and assembly of biomolecules [5–14]. However, despite its great biological relevance, our understanding of the microscopic mechanism of the dynamic motions of water molecules is much limited, although progress in time-resolved techniques and molecular simulations continually provide new knowledge in this field [15–21].

Recently, a large-amplitude angular jump mechanism shown in Fig. 1 was proposed to understand the reorientational motions of water molecules [22]. According to this mechanism, reorientation of water molecules occurs concertedly with simultaneous cleavage and formation of certain HBs, involving a large-amplitude angular jump, which is in contrast to the commonly accepted Debye diffusion picture [23]. This jump mechanism was further supported by recent experimental works using two-dimensional infrared spectroscopy [24,25]. Within the framework of the jump mechanism, the reorientational dynamics of water molecules in bulk water, aqueous solutions, and some confinement conditions have been extensively characterized both experimentally and computationally [26–30]. It is already clear that such jump motions are initiated by the fluctuations of local HB networks, e.g., the spontaneous occurrence of overcoordinated structural defects of the neighboring water molecules [22,30]. Because the jump motions can lead to perturbations to the HB networks of the surrounding water molecules due to the accompanying HB cleavage and formation, it is reasonable to anticipate that the jump of one water molecule can enhance the jump motions of the nearby water molecules, namely, the water jump motions are correlated. However, the correlation properties of such water reorientational jump motions remain elusive and have not been explicitly discussed. These sorts of correlated motions

may contribute to the experimentally observed cooperativity in many biological processes [9,31].

In fact, in Ref. [30], a memory effect was found in the waiting time between successive angular jumps based on molecular simulations, implying a certain correlation. In addition, Mazza *et al.* and Giovambattista *et al.* studied rotational dynamic heterogeneities in water by using molecular dynamics simulations [32,33], and found relations between rotational dynamic heterogeneities, which may suggest the spatial correlation between rotational motions. In a very recent work, Biman Jana *et al.* studied the propagation of the five-coordinated defect state at low temperature water [34], and found that the propagation was stringlike, with the length of propagation increasing with lowering temperature. All these studies inspired us to make a detailed characterization of the correlation properties of water angular jump motions on a molecular level.

In this paper, by using molecular dynamics (MD) simulations, we study the correlation between the large-amplitude angular jump motions in water molecules. We found that the reorientational jump of a certain water molecule can enhance the successive jump motions of the same water molecule and the surrounding water molecules. Such a correlation can extend to a distance of two water layers, and is propagated through the perturbations to the local HB networks. We also show a detailed molecular picture of the propagation of such jump-jump correlations.

II. METHODS

We used the software AMBER11 for MD simulations [35]. In the simulations, 8004 water molecules, described by the extended simple point charge (SPC-E) model [36], were placed in a cubic box with periodic boundary conditions. This water model has been shown to yield good agreement with experimental results [16,30]. The Ewald summation algorithm was used in calculating the long-range interactions [37], and the time step was set to 0.5 fs. The simulations were conducted at seven temperatures: 240, 260, 280, 290, 300, 320, and 340 K. Before the product simulations, the system was equilibrated in the *NPT* ensemble with the pressure of 1.0 atm. The product simulations were conducted in the microcanonical ensemble starting from the final structures of the *NPT* simulations. At

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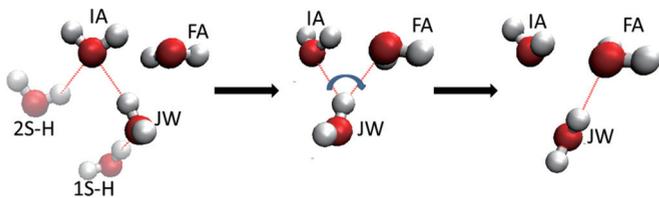


FIG. 1. (Color online) A schematic diagram of the water jump mechanism.

each temperature, the product simulation was conducted for more than 1 ns (from 1 ns at 340 K to 4 ns at 240 K). In the data analysis, we used the same criteria for HB definition as that in Ref. [22], namely, a HB is formed when the distance between the donor and acceptor oxygen atoms is less than 3.5 Å, and the angle formed by hydrogen-donor-acceptor atoms is less than 30.0° [22,38].

To identify the large-amplitude angular jump motions, we recorded all the events in which a water O-H breaks a HB and forms a new HB with another acceptor, and has a jump angle of more than 40° (see below for the definition of the jump angle). This jump amplitude was shown to be the lower limit for successful jump events due to steric hindrance between the initial and final water partners [30]. In order to exclude transient HB breaking events, we required that the newly formed HB has a lifetime of more than 150 fs, which was considered as the lower limit of HB lifetimes [19,39]. We tried other values of these two parameters and found that the results are robust [40]. We collected more than 3×10^6 successful jump events in our MD simulations at each temperature.

In characterizing the correlated motions, we focus on a number of water molecules, including the jumping water molecules (JW) that change their HB acceptors through large-amplitude angular jumps, the water molecules locating at the first coordination shell of the JW and forming HBs with the JW (denoted by 1S-H), and the water molecules locating at the second coordination shell of the JW and forming HBs with the 1S-H (denoted by 2S-H). See the schematic diagram of jump motions in Fig. 1 for more details of the definitions of these molecules. According to the above definitions, the initial HB acceptor (IA) and the final HB acceptor (FA), which forms an HB with JW before and after the jump of JW, respectively, are also classified as 1S-H. As a control, we also characterized the same properties for water molecules without jumping (UJW). UJW is defined as the molecules which do not jump within a time duration of a typical period of jump motions, i.e., 1.0 ps. The results have no drastic changes when we change the time duration from 1.0 to 2.0 ps. We emphasize that the HBs of UJW may break transiently during the 1.0 ps duration since transient HB breakings are not considered as successful jumps.

Following the work in Ref. [22], we used the angle θ , which is defined as the angle between the OH bond of JW and the bisector plane of angle $O_{IA} O_{JW} O_{FA}$, to describe the large-amplitude angular jump motions. During a successful jump event, the angle θ changes from $\sim -30^\circ$ to $\sim 30^\circ$ [22]. The jump angle was defined as the angle $O_{IA} O_{JW} O_{FA}$ when the jump occurs, the same as that defined in previous works [22,30]. By this definition we compared the initial average OH direction of the JW molecule, and the final average direction,

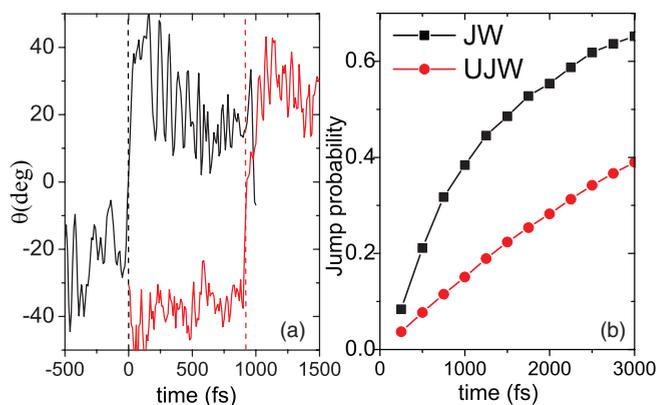


FIG. 2. (Color online) (a) A typical trajectory of successive jumps of JW. The black (left) dashed line is indicative of the time origin. (b) Probability of jump motions as a function of time for the water molecules JW and UJW, respectively, at 280 K.

thus eliminating the influence of the non-negligible amplitude of the librational motions of JW.

III. RESULTS AND DISCUSSIONS

First, we investigate the correlation between two successive large-amplitude angular jump motions of the JW molecule. Figure 2(a) shows a typical trajectory of successive jumps of JW. Note that the IA and FA used to calculate the rotation angle θ are different for the two successive jump transitions. We calculated the probability of successive jumps for the water molecules JW right after their previous jumps as a function of time at 280 K. The time point indicated by black (left) dashed line in Fig. 2(a) was set as the reference time for JW in Fig. 2(b). As a control, we also calculated the probability of jumps for the UJW molecules right after the 1.0 ps duration used for defining UJW. We can see from Fig. 2(b) that the water molecules JW show a much higher probability of jump motions right after their previous jumps compared to the water molecules UJW. Such results suggest that large-amplitude angular jump motion of a water molecule tends to enhance its subsequent jump motions, indicating the correlation of two successive jump events. Similar results are also observed at 290 and 300 K [40].

We then investigate the spatial correlation between jump events. For this purpose, we calculated the same jump probability for the water molecules surrounding JW and UJW, respectively, including the 1S-H and 2S-H molecules (see Sec. II for their definitions), and the results are shown in Fig. 3. For the 1S-H water molecules, we divided them into three groups: IA, FA, and other water molecules hydrogen bonded to JW. We can see that the water molecules surrounding JW have higher jump probabilities compared with their counterparts, i.e., the water molecules surrounding UJW [Fig. 3(a)]. This feature is particularly remarkable for the IA and FA molecules. All these results suggest that a jump of JW can enhance the jump motions of the surrounding water molecules, namely, the jump motions are correlated spatially.

Similar effects are observed for the 2S-H molecules, but the trend is much weaker, suggesting that the correlation can

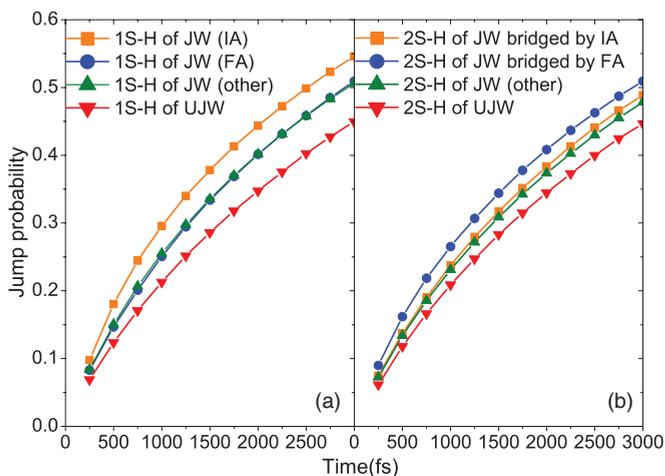


FIG. 3. (Color online) Probability of jump motions as a function of time for the water molecules (a) 1S-H and (b) 2S-H surrounding JW and UJW, respectively, at 280 K.

extend up to two water layers [Fig. 3(b)]. The results at 290 and 300 K are similar [40].

We also see from Figs. 2(b) and 3 that the correlation effects would vanish at long times, which may indicate that such a correlation has a lifetime. This result is consistent with the results in a previous work [30], where Laage and Hynes demonstrated the non-Markovian nature of the jump process at short delays, and found that there is a memory effect of jump events within approximately 2 ps. Here, from the rate of change of the jump probability of JW and UJW, we could estimate the lifetime of correlations to be about 1.5–2 ps, in good agreement with the suggestion in Ref. [30].

To quantitatively characterize the correlation length between the jump motions and its temperature dependence, we calculated the jump probabilities for the water molecules surrounding JW within the time duration of 1.0 ps right after the jump of the JW molecules as a function of their distances to the JW, and compared to those around UJW, at 240–340 K. The results at 240, 280, and 340 K are shown in Figs. 4(a)–4(c), respectively. We also show the radial distribution function (RDF) at 280 K in Fig. 4(d). We can see that, compared with water molecules around UJW, those around JW have a higher probability of jump motions at distances up to about 5–7 Å. Such differences again reflect the spatial correlation between the jumps of JW and its surrounding molecules. These results suggest that the correlation length of the water jump motions is around 5–7 Å (with a slight decrease with increasing temperature, as discussed below), which is in accord with the above discussions, namely, the jump of JW can increase the jump motions of the water molecules up to two water layers.

To more quantitatively characterize the temperature dependence of the correlation length, we defined the correlation length to be the distance at which the difference of jump probability between molecules around JW and UJW is less than 0.02, and the results are shown in Fig. 5. From Fig. 5 we see a slight decrease of the correlation length with increasing temperature, as expected. This should be due to the larger thermal fluctuation at higher temperatures, which would render the correlation effects less prominent.

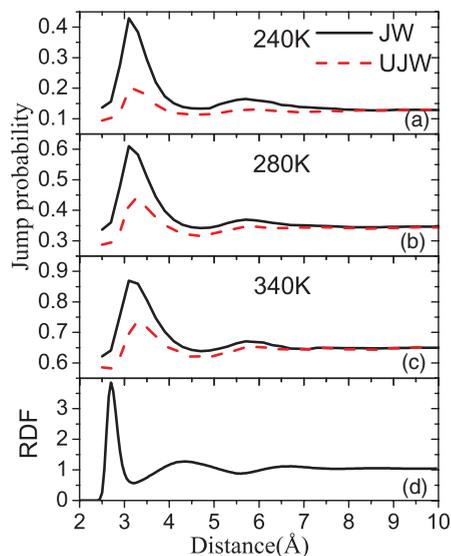


FIG. 4. (Color online) (a)–(c) Reorientational jump probability of water molecules in 1 ps as a function of distances to the central molecule, at 240, 280, and 340 K, respectively. (d) Radial distribution function of water at 280 K.

In addition, we can see that for both JW and UJW, the water molecules locating at the distances of ~ 3.5 and ~ 6.0 Å show higher jump probabilities. These peaks correspond to the minima of the water RDF [Fig. 4(d)] [41], where the local HB structure is less tetrahedral. This is consistent with the results in a recent work [42], where it was found that water molecules with a weakly structured environment show a higher jump rate. Thus, such a peak structure of the distributions of jump probabilities may suggest that the observed spatial correlation of the jump-jump motions is mediated by perturbations to the HB network.

To more quantitatively characterize the relation between the jump motions and the perturbations of the HB networks, we calculated $\langle N_{\max} - N_{\min} \rangle / \langle N \rangle$ as a measure of the perturbations to the local HB network. Here N denotes the number of HBs accepted or donated by certain molecules, including the IA, FA, and 2S-H water molecules hydrogen bonded to IA or FA, within the time duration of 1.0 ps right after the jump of JW. N_{\max} and N_{\min} are the maximum and

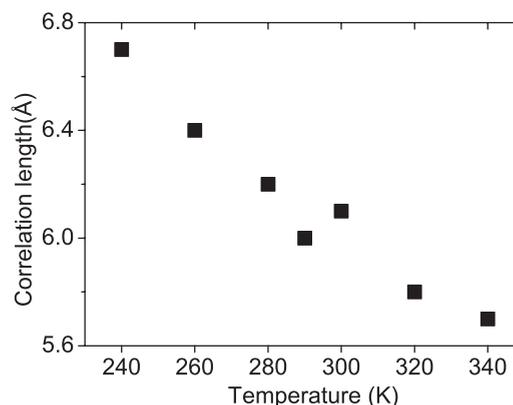


FIG. 5. Temperature dependence of the correlation length of water jump motions (defined in the text).

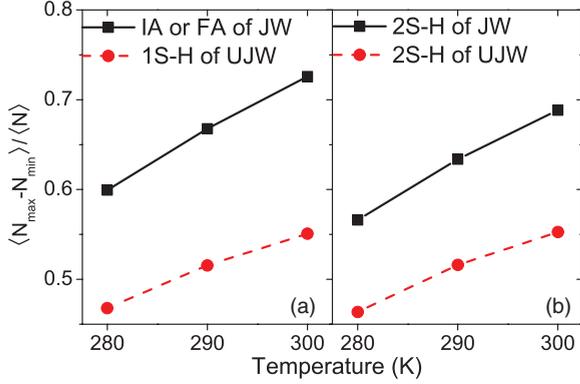


FIG. 6. (Color online) (a) Fluctuation of HB networks at different temperatures for IA or FA of JW (black solid line), and 1S-H of UJW (red dashed line). (b) Fluctuation of HB networks at different temperatures for 2S-H of JW hydrogen bonded to IA or FA (black solid line), and 2S-H of UJW (red dashed line).

minimum of N , and $\langle \dots \rangle$ stands for the average over time and water molecules. Figure 6 shows $\langle N_{\max} - N_{\min} \rangle / \langle N \rangle$ at different temperatures for the IA and FA molecules [Fig. 6(a)], and for the 2S-H water molecules hydrogen bonded to IA or FA [Fig. 6(b)]. As a control, we also calculated the same properties for the 1S-H and 2S-H of the UJW molecules, within the time duration of 1.0 ps after the time duration of 1.0 ps used for defining UJW. It is quite clear that at each temperature, IA and FA, as well as their neighbors, show much higher values of $\langle N_{\max} - N_{\min} \rangle / \langle N \rangle$, indicating larger fluctuations of the HB network, compared with the neighbors of the water molecules without jumps. Such results further support the above discussions that the correlation of water jump motions is propagated through the perturbations to the local HB networks. The large-amplitude angular jump of JW induces larger fluctuations of the HB networks of the nearby water molecules, which leads to enhanced jump motions of the same water molecules and the nearby water molecules during the subsequent time duration, as observed in this work.

It was shown that the free-energy cost for the fluctuation in the hydrogen-bond network preceding a jump event is below the thermal energy $k_B T$ [30], where k_B is the Boltzmann constant. Consequently, spontaneous thermal fluctuation can contribute to the perturbations of the HB network. From Fig. 6, we can see that even for the water molecules around the UJW, $\langle N_{\max} - N_{\min} \rangle / \langle N \rangle$ has considerable values, which may suggest the contributions of thermal fluctuations. Nevertheless, in the above discussions, we are interested in the differences of the HB network fluctuations around JW and UJW, which reflect the effects of jumping events.

The above results clearly demonstrate that the jump of JW enhances the fluctuations of the nearby HB network and the jump motions of the surrounding water. It is important to show how the fluctuations of the HB network and jump motions are correlated at a single molecular level. For this purpose, we monitored all the JW jump events which are accompanied by the jump of surrounding water molecules, and analyzed the variation of the HB network. We show the trajectory of one such event in Fig. 7 to illustrate the molecular picture of the jump correlations observed in this work. The key

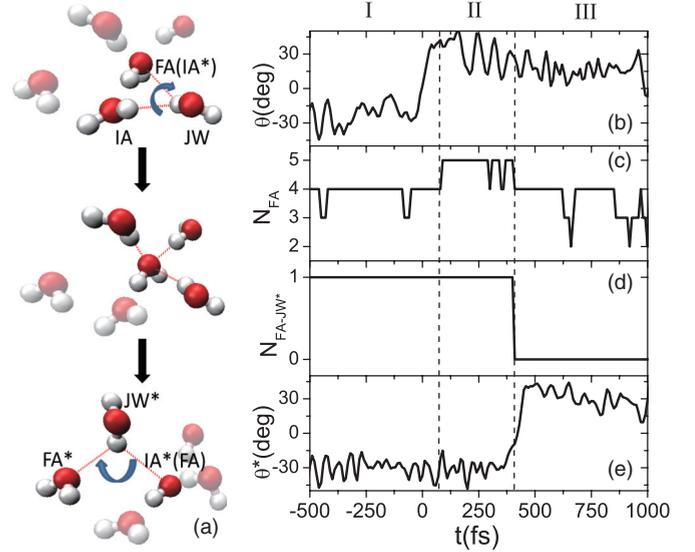


FIG. 7. (Color online) (a) Snapshots in a trajectory of two correlated jump events. Evolution of (b) θ , (c) N_{FA} , (d) N_{FA-JW^*} , and (e) θ^* with time corresponding to the trajectory shown in (a). θ and θ^* are the rotation angles of the jumps of JW and JW*, respectively. N_{FA} is the number of HBs formed by the FA molecule, and N_{FA-JW^*} is the number of HBs formed between FA and JW*.

snapshots are shown in Fig. 7(a) for a better understanding. In this trajectory, first, JW jumps from IA to FA [Fig. 7(b)], making FA transiently five-coordinated [Fig. 7(c)]. Due to the metastable nature of the overcoordinated HB structure, one of the water molecules (JW*) coordinated to FA breaks the HB [Fig. 7(d)] and forms a new one with another water molecule (FA*), by a large-amplitude angular jump motion [Fig. 7(e)]. The above trajectory illustrates how the large-amplitude jump of JW induces the perturbation of the HB network involving FA, which in turn leads to the jump of JW*.

The above analysis of the trajectory of the pair of jumps shown in Fig. 7 suggests that the trajectory of successive jumps can be separated into three stages: (I) prejump stage of JW; (II) transient stage; and (III) postjump stage of JW* [Figs. 7(b)–7(e)]. The rotation angles of JW and JW* (denoted by θ and θ^* , respectively) are similar in stages I and III, but have a large difference in stage II. Therefore, we used the difference of rotation angles $\Delta\theta$ to represent the three stages. Specifically, the trajectories are in stages I or III (I&III) when $|\Delta\theta| < 10^\circ$, and stage II when $|\Delta\theta| > 40^\circ$, respectively.

We next show that the above molecular mechanism of spatial correlation between jump motions is generic. For this purpose, we calculated the distributions of the number of HBs coordinated to the FA molecules for all the collected trajectories of successive jumps. The results are shown in Fig. 8 for the snapshots with $|\Delta\theta| < 10^\circ$ (black) and $|\Delta\theta| > 40^\circ$ (red, light gray) at temperatures of (a) 280, (b) 290, and (c) 300 K, respectively. We can see that the distributions have significant differences in stages II and I&III. In stage I&III, the coordination number of FA is dominated by four, which represents regular HB network. In comparison, in stage II, the five-coordinated HB network of FA, which represents perturbations to the regular HB network, has a significant population ($\sim 30\%$). Such a statistical significance of the metastable state

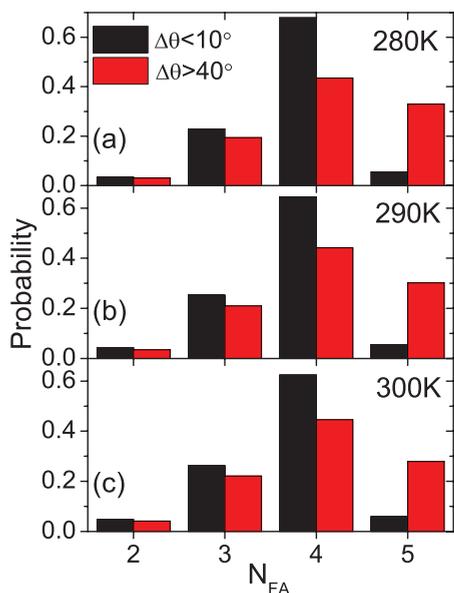


FIG. 8. (Color online) Distributions of the coordination number of the molecule FA in stage I&III and stage II at different temperatures. The difference of rotation angles $\Delta\theta$ was used to represent different stages. The trajectories are in stage I or III (I&III) when $|\Delta\theta| < 10^\circ$, and stage II when $|\Delta\theta| > 40^\circ$, respectively.

of the FA molecules in the transient stage clearly supports that the “JW jump \rightarrow FA overcoordination \rightarrow JW* jump” pathway is generic, and demonstrates that the spatial correlation of water jump motions is largely propagated through the perturbations to the local HB networks.

Laage and Hynes recently investigated the average path of jump events and found that fluctuation in the HB network would facilitate the motion of the O-H of JW away from the

overcoordinated IA and toward the undercoordinated FA in a jump event [30]. Here we further demonstrate the relation between the correlations of jump events and the perturbations to the HB network. The “JW jump \rightarrow FA overcoordination \rightarrow JW* jump” pathway presented here is consistent with the stringlike propagation of the five-coordinated defect state proposed by Biman Jana *et al.* in studying the dynamics and thermodynamic anomalies of water molecules at low temperatures [34]. Differently, here we mainly discuss the temporal and spatial correlations of the large-amplitude jump rotations.

IV. CONCLUSION

The recently revealed large-amplitude jump mechanism of water reorientation inspired us to investigate the correlations between the jump motions. Our results demonstrate that the large-angular jump motions of a water molecule can enhance the successive jump motions of the same water molecule and the surrounding water molecules. Such a correlation can extend up to a distance of two water layers and is propagated through the perturbations to the local HB networks. These correlation features of the water dynamics may be useful for understanding some biological processes involving the motions of water molecules.

ACKNOWLEDGMENTS

The authors thank the Shanghai Supercomputer Center and High Performance Computing Center of Nanjing University for the computational support. This work was supported by the National Natural Science Foundation of China Grant Nos. (11174134, 91127026, and 81121062), Natural Science Foundation of Jiangsu Province (Grant No. BK2011546), the 973 program (2013CB834100), and PAPD.

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