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Dynamics of supercooled water in nanotubes: Cage correlation function and diffusion coefficient

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Dynamics of low-temperature water in nanostructured materials is important to a variety of phenomena, ranging from transport in cement and asphaltene, to conformational dynamics of proteins in "crowded" cellular environments, survival of microorganisms at very low temperatures, and diffusion in nanogeoscience. Using silicon-carbide nanotubes as a prototype of nanostructured materials, extensive molecular dynamics simulations were carried out to study the cage correlation function $C(t)$ and self-diffusivity D of supercooled water in the nanotubes. $C(t)$, which measures changes in the atomic surroundings inside the nanotube, follows the Kohlrausch-Williams-Watts law, $C(t) \sim \exp[-(t/\tau)^{\beta}]$, where τ is a relaxation time and β is a topological exponent. For the temperature range 220 K $< T \leqslant$ 273 K, we find $\beta \simeq 0.438$, in excellent agreement with and confirming the prediction by Phillips [Rep. Prog. Phys. **59**, 1133 (1996)], $\beta = 3/7$. The self-diffusivity manifests a transition around 230 K, very close to 228 K, the temperature at which a fragile-to-strong dynamic crossover is supposed to happen. Thus the results indicate that water does not freeze in the nanotube over the studied temperature range, and that the Stokes-Einstein relation breaks down.

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I. INTRODUCTION

Water, in addition to being a fundamental ingredient of life, has many unusual properties [1], such as the lowest density at 4 °C, as well as its coexistence within two distinct forms—polymorphism [2]—and the abnormal behavior of its isothermal compressibility, isobaric heat capacity, and thermal expansion coefficient at temperatures between homogeneous nucleation (231 K) and its melting point of 273 K, all of which have been studied intensively. In addition, water can be in a supercooled state between 273 K and about 230 K [1]. The supercooled liquid state is important from a biological standpoint because it can preserve microorganism during freezing. In contrast to other liquids, at 273 K and lower temperatures, water volumetric expansion is associated with reduction in the entropy due to the tetrahedral symmetry of the local order around each water molecule [1], related to hydrogen bonding. Under bulk conditions three forms of glassy water exist, namely, low-density amorphous (LDA) ice, and high-density and very high-density amorphous (HDA and VHDA, respectively) ice. The region between homogeneous nucleation temperature (231 K) and crystallization temperature is above the glass transition temperature of 165 K. The gradual crystallization process by decreasing the temperature has been explained by the hypothesis that there exists a liquid-liquid critical point, based on structural changes governed by hydrogen bonds during water clustering process, and the development of a tetrahedral-coordinated network. The nature of water at 228 K, which has been hypothesized to be a second, low-temperature critical point of water, is still under investigation.

Even more interesting and important phenomena occur when water is in confined media. Understanding various properties of water in such media is highly important to many physical, chemical, and biological phenomena, as they appear to be fundamentally different from their bulk counterparts. For example, diffusion of water in nanopores and nanotubes is governed by a process that is different from the bulk phase [3–5], because while diffusion under bulk conditions follows the Einstein relation and only local temperature and pressure influence the transport process, the same is not true about water diffusion in a confined medium. In such a medium the interactions of water molecules with the solid walls, which are mainly of the van der Waals and Coulombic type, affect their mobility and usually reduce the rate of molecular diffusion [6,7]. Moreover, a water molecule in the bulk is, on average, hydrogen-bonded to four neighboring molecules. If one hydrogen bond breaks, the O-H-O configuration moves away from linearity by more than 25°, and as three or four H-bonds are broken, the molecules undergo rotational diffusion. This is not, however, the case in a confined medium. Thus, despite considerable experimental and theoretical-computational works to understand the possible deviations of the behavior of water molecules in confined media from that in the bulk, no reasonably complete understanding of the properties of water in confined media has emerged. For example, it is not yet clear whether the Stokes-Einstein relation is followed by water molecules in nanotubes.

One way of achieving subcooling is [8] to use pores larger than a critical size of 3–4 nm, in which water freezes to a mixture of cubic crystals and amorphous ice. In smaller pores, however, water does not crystallize. Another possibility is to study the behavior of water in nanotubes. Kolesnikov *et al.* [9] investigated the behavior of encapsulated water inside carbon nanotubes (CNTs), and reported an anomalous "soft" dynamics characterized by pliable hydrogen bonds, anharmonic intermolecular potentials, and large-amplitude motions. They proposed that the structure of water in nanotubes consists of a square-ice sheet wrapped into a cylinder inside the CNT with the interior molecules being in a chainlike configuration, and that the motion of the water molecules is enhanced along the chains. Such an enhanced motion well below the freezing point has been verified experimentally.

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A key characteristic of water inside narrow nanotubes may be fast formation of hydrogen bonds that is sustainable and lasts longer than those in the bulk. Based on molecular dynamics (MD) simulations, Mashl *et al.* [10] proposed 8.6 Å as the critical radius of the CNTs in which the water displays anomalous icelike behavior in both symmetry and mobility, while retaining a degree of resemblance to liquid water, which is hydrogen bonding. Thus, in confined media such as nanotubes, water manifests a behavior that is intermediate between solid and liquid states. The calculated mean-square displacements (MSDs) of the water molecules inside the CNTs are consistently smaller than those in the bulk. But, chainlike water molecules travel up to 40 times faster inside CNTs than in the bulk, hence providing excellent channels for molecular transfer [11–13].

Using MD simulation, we study in this paper the dynamics of water motion at low temperatures in a relatively new nanostructured material, namely, silicon-carbide nanotubes (SiCNTs). In particular, we study the so-called *cage effect* on water in the SiCNTs, and compute water's self-diffusivity. Many liquids, when cooled rapidly below their melting point, undergo a glass transition, causing their viscosity to increase dramatically. The increase is due to a slowdown of the dynamics often referred to as the cage effect, rather than any significant structural changes: confinement of any of the liquid's molecules by a "cage" formed by its neighboring molecules. When the cage rearranges, the liquid relaxes and the molecules diffuse in the system [14–18]. The rate of change of molecular surroundings of a molecule is characterized [18] by a cage correlation (CC) function. Over time, the CC function decays, characterizing the anomalous slowing of the dynamics (see below). While the dynamics of the cages in liquids under bulk conditions have been studied both experimentally and by computer simulations [19–26], it has not, to our knowledge, been studied in a nanostructured material, such as a nanotube that we study in this paper. In addition to its fundamental scientific importance, the phenomenon that we study is relevant to the behavior of water in biological materials in, for example, confined or "crowded" cellular environments.

The rest of this paper is organized as follows. In the next section we describe the nanotubes that we employ in the study. Section III provides the details of the MD simulation, while the results are presented and discussed in Sec. IV. The paper is summarized in Sec. V.

II. SILICON-CARBIDE NANOTUBES

Silicon carbide is, of course, an important material with many excellent properties, such as high fracture toughness, thermal shock resistance, and the ability for withstanding high temperatures and corrosive environments. It is also a promising material for fabrication of various nanostructured materials, such as nanoporous membranes [27,28] with pore sizes on the order of a few angstroms that are used for separation of fluid mixtures in harsh environments in which other types of membranes fail.

We choose to work with the SiCNTs due to their rich behavior. The tubules formation of the SiCNTs, and the existence of specific charge arrangement on their surface make them attractive materials for a variety of purposes. Moreover, the polar-polar repulsion between the π bonds of SiC and water does *not* allow water molecules to come too close to the surface, hence acting effectively as a "nonwetting" surface (to the extent that the concept of wetting can be considered in such nanostructured materials).

Stable SiCNTs have a one-to-one ratio of Si and C, while any other Si-to-C ratio causes their collapse onto a nanowire or clusters with solid interior [29]. We have recently fabricated the SiCNTs [30] and nanofibers made of SiC [31] and, using MD simulations, have studied [32,33] a variety of phenomena in them, both in the gas and liquid phases. In particular, our recent MD simulation [33] of flow of water in the SiCNTs at room temperature indicated that, for a given volume flow rate of water, the pressure drop needed to induce water flow in SiCNTs is at least one order of magnitude smaller than their CNT counterparts.

Similar to CNTs, there are three types of SiCNTs, but in this paper we work with the zigzag-type nanotubes, denoted by $(m,0)$, in which one has rows of alternating Si and C atoms perpendicular to the tube's axis. Thus, in the honeycomb lattice of SiC—its primary structure—each Si has three C neighbors and vice versa, and only Si-C bonds are formed. Such nanotubes may be formed by simply rolling the lattice of Si and C atoms. The length of the Si-C bond is about 1.8 Å, 10% longer than that of the C-C bond in CNTs.

III. MOLECULAR DYNAMICS SIMULATION

To carry out the MD simulations, we first generated the SiCNT using energy minimization and an enhanced algorithm for the Newton's method, together with the conjugate-gradient minimization. The simulations were carried out in the canonical (NVT) ensemble. The force field used was condensed-phase optimized molecular potentials for atomistic simulation studies (COMPASS). The Nosé-Hoover-Langevin dynamics was used for the thermostat, and the Ewald summation was utilized for the calculation of the electrostatic and long-range interactions. The generated (12,0) SiCNT with a diameter of 11.9 Å was anchored and immersed in bulk water; see Fig. 1. The water molecules were represented by the extended simple point charge model (the SPC/E model) [34]. After the tube was immersed in water, energy minimization was used again. Multiple runs were then carried out in the range 200–300 K, in order to evaluate the CC function by monitoring the movement of water molecules inside the nanotube, and compute the self-diffusivity of water. The simulations were carried out for at least 1000 ps, analyzing the results in 1 ps increments.

To calculate the CC function we used a generalized neighbor list [35] to keep track of each atom's neighbors. In liquids, such as water, the immediate neighborhood of an atom is best described by a list of other atoms that make up the first solvation shell. If the list of an atom's neighbors at time t is identical to the list at time $t = 0$, the CC function takes on a value of 1 for that atom. If, however, any of the original neighboring molecules are not in the cage at time t, the implication is that the atom (or the molecule) has hopped outside the cage and, thus, the CC function is zero at that time. Thus, if we average over all the atoms in the simulation, the CC function provides us with a direct way of measuring the hopping times even from very short MD simulations.

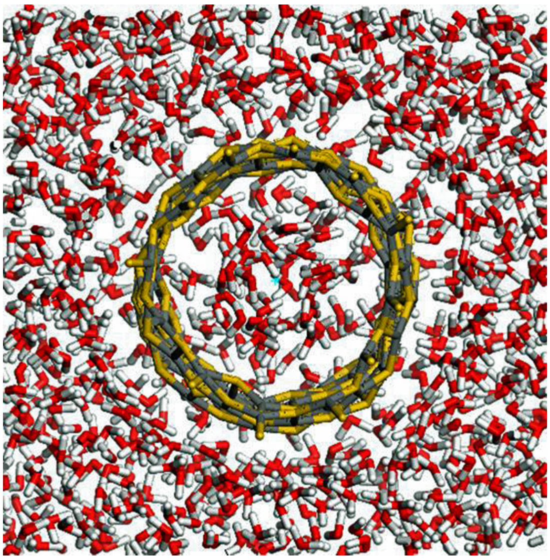

FIG. 1. (Color online) Zigzag SiCNT immersed in water.

We used [35] the location of the oxygen in water as the basis for calculating the CC function, as it is the closest to the center of mass of the water molecules. By considering how each atom's location varies with time, we generated a list in the form of a generalized vector of neighbors for each individual oxygen atom i of the water molecules inside the nanotube with radial distance r_{list},

$$\mathbf{L}_i(t) = [f(r_{ij})], \tag{1}$$

with $j = 1,2,\ldots,N$, where N is the total number of molecules (atoms) in the system, and

$$f(r_{ij}) = \begin{cases} 1, & r_{ij} \leqslant r_{\text{list}}, \\ 0, & \text{otherwise}. \end{cases} \tag{2}$$

There are various ways of selecting r_{list} [35], depending on how much computer time one can afford. Given a suitable r_{list}, the CC function at time t is given by

$$C(t) = \frac{\langle \mathbf{L}_i(0)\cdot\mathbf{L}_i(t)\rangle}{\langle \mathbf{L}_i^2(0)\rangle}. \tag{3}$$

Note that, since even in the bulk the neighborhood of molecules must change completely in order for $C(t)$ to vanish, its decay with time is slow. Clearly, then, in a nanostructured material, the decay of $C(t)$ is even slower.

IV. RESULTS AND DISCUSSION

We calculated two important properties of the supercooled water in the SiCNT, namely, the CC function and the diffusion coefficient of water. Both were computed over a wide range of temperature. In what follows we present the results and discuss their implications.

A. Cage correlation function

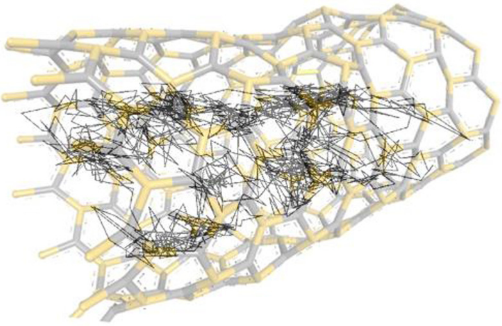

FIG. 2. (Color online) Diffusion paths of the water molecules inside the nanotube at a low temperature.

Figure 2 presents a sample of the diffusion paths for the water molecules inside the nanotube, indicating a complex ensemble. Figure 3 presents the time dependence of $C(t)$ at 250 K and 273 K. The decay of $C(t)$ at 273 K is faster than at 250 K, as expected, but is still slow. Our MD simulation at 220 K and lower temperatures indicated no significant decay of $C(t)$ over the simulation time that we could afford. The decay is simply too slow and computing $C(t)$ at such temperatures requires much longer simulations. The slow decay of $C(t)$ has motivated its representation by the Kohlrausch-Williams-Watts [36,37] stretched exponential form,

$$C(t) \propto \exp[-(t/\tau)^{\beta}], \tag{4}$$

where τ is a relaxation time scale, and β is a shape factor that has to do with the topology of the space in which the molecules move. In general [38,39],

$$\beta = \frac{d^*}{d^*+2}, \tag{5}$$

where d^* is an *effective* dimensionality of the system. It has been proposed that if only short-range forces are present, then $d^* = 3$, so that $\beta = 3/5$. Phillips proposed [40], however, that in the presence of both short- and long-range forces, $d^* = d/2$, where d is the spatial dimension of the system, so that $\beta = d/(d+4)$. With $d = 3$, one obtains $\beta = 3/7 \approx 0.43$. To our knowledge, confirmation of Phillips' prediction for any liquid with short- and long-range forces in any system has not been reported previously, particularly in a confined medium such as a nanotube.

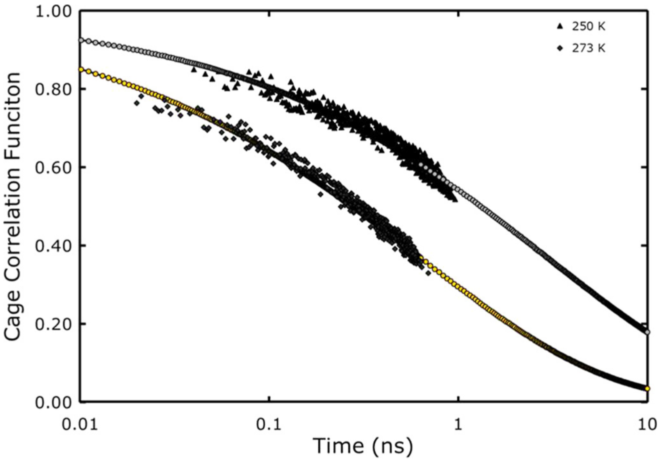

FIG. 3. (Color online) Cage correlation function at two temperatures.

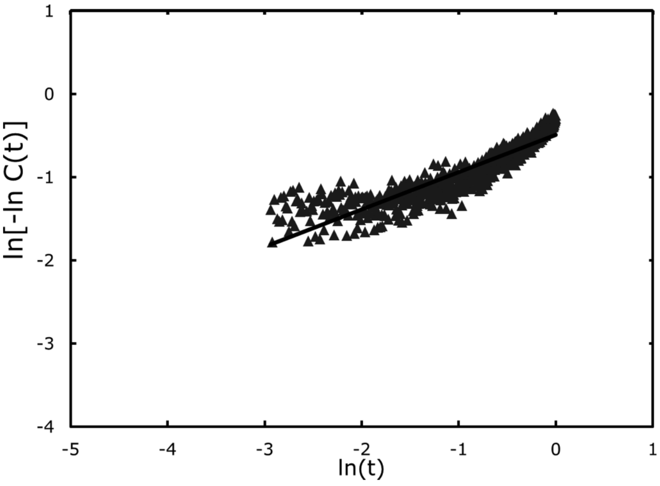

FIG. 4. Double logarithmic plot of the cage correlation function at 250 K.

To estimate the exponent β, we rewrite Eq. (4) as $\ln[-\ln C(t)] \propto \beta(\ln t - \ln \tau)$. Thus a double logarithmic plot of $C(t)$ versus $\ln t$ must be a straight line with a slope β. Figures 4 and 5 present such plots at 250 K and 273 K. Both yield $\beta \simeq 0.438$, in excellent agreement with Phillips' theoretical prediction, $\beta = 3/7$. We also obtained the estimates for the relaxation times, $\tau \simeq 3.0$ ns and 0.64 ns at 250 K and 273 K, respectively. Thus τ increases significantly as the system is cooled down.

Our estimate of the exponent β should be compared with those obtained under the bulk condition at low temperatures. For Lennard-Jones fluids the estimate $\beta \approx 1/2$ has been reported [35], although values as high as $\beta \approx 0.8$ have also been reported [41]. Phillips [40] described a variety of supercooled liquids for which the value of β is around 3/5. Note also that if we plot $C(t)$ versus t/τ, the CC functions at various temperatures should collapse onto each other. This is shown in Fig. 6, and confirms the data collapse.

The CC function represents an average over all the atoms in the simulation. As such, it is not capable of distinguishing between nonstretched exponential decay, which is caused by a static distribution of local environments in the nanotube, and nonstretched exponential decrease due to dynamically fluctuating local environments. However, because

$$C(t) = \int_0^\infty \rho(x)\exp(-xt)dx, \qquad (6)$$

where $\rho(x)$ is the distribution of hopping rates of the molecules into and out of the cages, the CC function does provide information on $\rho(x)$.

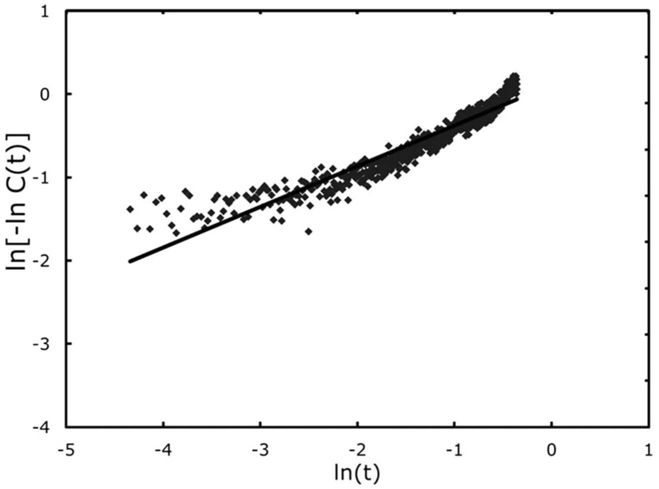

FIG. 5. Same as in Fig. 4, but at 273 K.

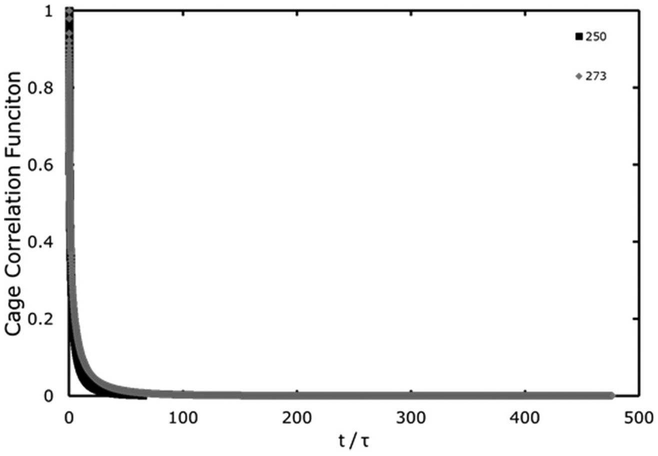

FIG. 6. Cage correlation function $C(t)$ vs t/τ, where τ is the relaxation time.

B. Self-diffusivity

Since the computed CC function did not take on a value of 1 at all times that would have indicated a frozen state, we also computed the self-diffusivity of the water molecules in the nanotube by calculating their mean-square displacements. The results are presented in Fig. 7 where we show a log-linear plot of $1/D$ versus $1000/T$. There are clearly two distinct regimes, and a crossover at around $T = 230$ K. The transition

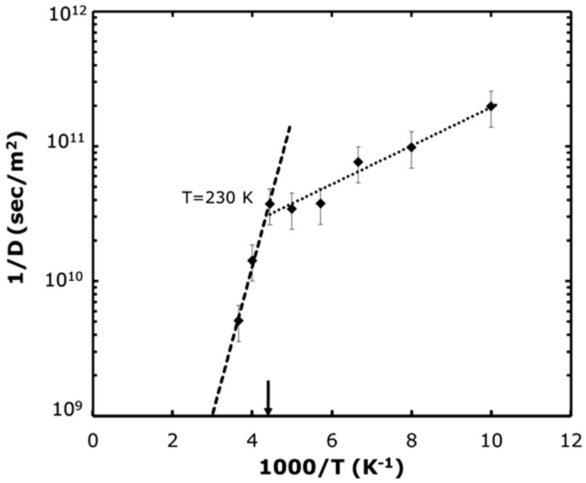

FIG. 7. Log-linear plot of the inverse of the self-diffusivity D of water vs the inverse of temperature T. The arrow indicates the location of a fragile-to-strong transition.

temperature is in agreement with the proposal of Ito *et al.* [42] that there should be a transition at 228 K from a "fragile" state at room and moderately supercooled temperatures to a "strong" liquid state. Below 230 K the values of the diffusivity are low, but beyond the crossover point they sharply increase as T approaches the bulk melting point 273 K.

C. Breakdown of the Stokes-Einstein relation

The fact that the CC function is described by a stretched exponential function implies that diffusion of the water molecules inside the nanotubes does *not* follow the Stokes-Einstein (SE) relation between the self-diffusivity and the viscosity. Indeed, if we assume that the viscosity is proportional to the relaxation time, then [3,4] a plot of $D\tau/T$ versus T must be a horizontal line and constant, if the Stokes-Einstein relation holds. Our extensive data for D and limited data for τ indicate otherwise. The plot yields a nonconstant curve, indicating the breakdown of the SE relation. We are currently computing the water viscosity within the SiCNTs to test the hypothesis more accurately.

V. SUMMARY AND CONCLUSIONS

Using extensive MD simulation, we studied dynamics of water motion in SiCNTs. The results confirm the stretched-exponential nature of the cage correlation function with a topological exponent β that is in complete agreement with Phillips' prediction [40], $\beta = 3/7$. We also computed the self-diffusivity of water molecules in the nanotube. The results indicate a transition from a fragile to a strong liquid state at 230 K.

The results have important fundamental and practical applications and implications, particularly to biology. First, that water does not freeze in the nanotube below its bulk freezing temperature indicates that such nanostructured materials may be ideal for, for example, preserving DNA samples and other biological tissues, rather than the common practice of freezing them. Second, water not freezing in such nanostructured materials provides a plausible explanation for why microorganisms survive at very low temperatures whereas, for example, human beings do not. Third, the fact that the Stokes-Einstein relation breaks down in the nanotube is significant, highlighting another important difference between the state of liquids in the bulk and in confined media. Fourth, the transition in the self-diffusivity indicates a structural change between two states of water, fragile and strong.

Both the breakdown of the Stokes-Einstein relation and the transition in the self-diffusivity add to the list of other unusual properties of water, but from the point of view of the transport properties of water. Finally, the results are also relevant to a highly practical, everyday problem, namely, transport of water in cements and asphalts, both of which are damaged by it, particularly at extreme conditions. The results open up a venue for understanding this critical problem, and addressing it.

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