Lifshitz interaction can promote ice growth at water-silica interfaces

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At air-water interfaces, the Lifshitz interaction by itself does not promote ice growth. On the contrary, we find that the Lifshitz force promotes the growth of an ice film, up to 1–8 nm thickness, near silica-water interfaces at the triple point of water. This is achieved in a system where the combined effect of the retardation and the zero frequency mode influences the short-range interactions at low temperatures, contrary to common understanding. Cancellation between the positive and negative contributions in the Lifshitz spectral function is reversed in silica with high porosity. Our results provide a model for how water freezes on glass and other surfaces.

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Although water in its different forms has been studied for a very long time, several properties of water and ice remain uncertain and are currently under intense investigation [1–4]. The question we want to address in the present paper is to what extent the fluctuation-induced Lifshitz interaction can promote the growth of ice films at water-solid interfaces, at the triple point of water. Particles and surfaces, e.g., quartz, soot, or bacteria, in supercooled water are known experimentally to nucleate ice formation [5–7]. Here, we focus on interfaces between water and silica-based materials and examine the roles of several intervening factors in the sum over frequency modes (Matsubara terms) contributing to the Lifshitz free energy.

Quantum fluctuations in the electromagnetic field result in van der Waals interactions, which in their unretarded form were explained by London in terms of frequency-dependent responses to the fluctuations in the polarizable atoms constituting the material medium [8]. The understanding of these interactions was revolutionized when Casimir introduced retardation effects [9]. The theory was later generalized by Lifshitz to include dielectric materials [10,11]. The Lifshitz formula in Eq. (1), derived for three-layer planar geometries [11], gives the interaction energy between two semi-infinite dielectric media described by their frequency-dependent dielectric permittivities as well as the dielectric permittivity of the medium separating them (see Fig. 1).

The purpose of the present work is twofold. First, we want to show that a finite size ice film, nucleated by a solid-water interface, can be energetically favorable even when only the Lifshitz interaction is accounted for. Second, we want to highlight a relevant contribution from the zero frequency term in the expression for the Lifshitz energy in a region where it is not expected to be important. The temperature dependence of the Casimir force between metal surfaces [11–14] relies strongly on the exact behavior of the low-frequency dielectric function of metals. These and many other investigations have provided support for the notion that the zero frequency term would only be relevant at high temperatures or large surface separations at a moderate temperature. In biological systems that involve water, the zero frequency term contributes substantially to the total Lifshitz interaction energy because of the high static dielectric permittivity of the water compared to the interacting media [15,16].

In this paper, we will show that for three-layer planar geometries, where an attractive-repulsive force transition can occur, it is possible to find systems in which the combined effect of retardation and the zero frequency term determines what happens with the interaction across extremely thin sheets. The mechanism behind this is a cancellation between the positive (repulsive) and negative (attractive) contributions from the different frequency regions, which leads to a diminished contribution from the nonzero Matsubara terms and thus renders the zero frequency Matsubara term dominant.

We emphasize that the system, in spite of its apparent simplicity, is far from trivial. The resulting value of the Lifshitz energy is dependent on an interplay between different factors:

(i) The crossing in the curves for the permittivities ε as functions of the imaginary frequency ζ, where the crossing occurs in the optical region, results in a switch from attractive to repulsive contributions to the Lifshitz force.

(ii) The need to include retardation effects in the formalism: This may appear surprising, as retardation effects due to the finite speed of light c are usually related to cases where the gap widths are large.

(iii) The dominant role played by the zero frequency Matsubara term $n = 0$, which is a direct consequence of the aforementioned two factors: This may also be somewhat unexpected, in view of the circumstance that the $n = 0$ term is usually taken to be important only in the limits of large separation distance L at a moderate temperature

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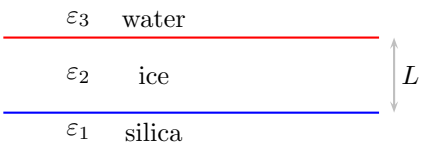

FIG. 1. Ice (ε_2) of thickness L at the interface of water (ε_3) and silica (ε_1), illustrated here as three planar regions of infinite extent.

T, or high temperature at moderate separation. (Observe that in the special case of a nondispersive medium, the single nondimensional parameter of importance in the Lifshitz integral is $Lk_BT/\hbar c$.)

The need to include all these effects stems of course from the complicated Lifshitz sum-integral, when the dispersive properties of the material components are accounted for accurately. In a three-layer planar system, where medium 1 is interacting with medium 3 across medium 2, the system tries to minimize the interaction energy, which manifests as a force of attraction if $[\varepsilon_1(i\zeta)-\varepsilon_2(i\zeta)][\varepsilon_3(i\zeta)-\varepsilon_2(i\zeta)]>0$ and a force of repulsion for $[\varepsilon_1(i\zeta)-\varepsilon_2(i\zeta)][\varepsilon_3(i\zeta)-\varepsilon_2(i\zeta)]<0$. These conditions for attraction and repulsion must hold over a wide frequency range because they occur within the Lifshitz sum-integral. The plausibility of the repulsive Lifshitz force between two dielectric objects with an intervening medium of suitable dielectric permittivity was first discussed by Dzyaloshinskii *et al.* [11] and has been observed experimentally [17–21]. Earlier experimental and theoretical studies are comprehensively discussed in Ref. [22]. Elbaum and Schick observed that the difference between the dielectric permittivities of ice and water changes sign at the transition frequency ($\zeta_a \approx 1.60\times10^{16}$ rad/s), as shown in Fig. 2 [23]. Thus, the contribution to the Lifshitz force, above and below the transition frequency ζ_a, is attractive and repulsive in nature, respectively. Furthermore, the difference between the dielectric permittivities of ice and water changes sign again at frequencies lower than the first Matsubara frequency, thus affecting the overall behavior of the Lifshitz force. Elbaum and Schick showed that these attractive and repulsive contributions for the ice-water-vapor system, at the triple point, lead to the formation of a thin layer of water at the interface of ice and vapor [23]. The scale for the thickness of the layer of water is set by the transition distance c/ζ_a. Most often it is argued that the retardation effects can be neglected if the distance is less than a few tens of nanometers. However, several studies [23,26–29] highlight the importance of including the retardation effect even at separation distances of less than 10 nm.

We investigate if a thin layer of ice at the interface of silica and water will grow (freeze) or vanish (melt), near the triple point of water, assisted exclusively by the Lifshitz interaction. In Ref. [30], Elbaum and Schick find that a thin sheet of ice does not grow at the water-vapor interface. In contrast, we report that the Lifshitz force does assist ice growth at the silica-water interface. The thickness of the ice layer formed at the silica-water interface varies with the permittivities of the silica substrate. (In Ref. [31], Dash *et al.* thoroughly reviewed a related phenomenon of the premelting of ice, which was also considered by some of us in Ref. [32], where we showed that it is essential to have a vapor layer between ice and a silica surface to have premelting of the ice.)

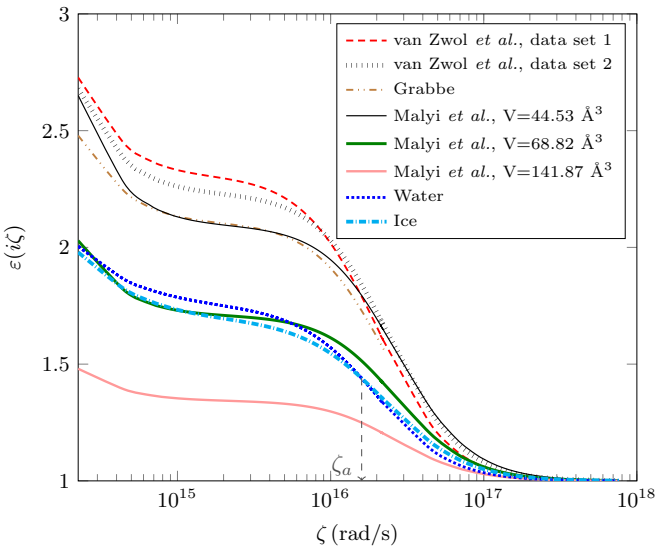

FIG. 2. Permittivity as a function of frequency for ice, water, and different silica materials. The static values $\varepsilon(0)$ for ice and water are 91.5 and 88.2, respectively, using data from Elbaum and Schick [23]. For different SiO_2 materials, the static values are 3.90, 2.62, and 1.69 using data from Malyi *et al.* [24] for volumes 44.53, 68.82, and 141.87 Å^3, respectively (here extended to include phonon contributions), 3.80 from Grabbe [25], and 3.90 from data set 1 and data set 2 of van Zwol and Palasantzas [26]. The transition frequency, $\zeta_a \approx 1.60\times10^{16}$ rad/s, is where the permittivities of ice and water cross in the optical frequency region.

To study ice growth at the silica-water interface, we consider a model system with a planar silica surface interacting with water across a thin planar ice film of thickness L, as illustrated in Fig. 1. The ice sheet thicknesses that we discuss are typically in the range 1–8 nm. Recently, Schlaich *et al.* [33] showed that the dielectric functions for films thicker than 1 nm approached their bulk values. Thus, to predict trends, it should be sufficient to use bulk dielectric functions for the thin ice layer. The Lifshitz interaction free energy per unit area F is expressed as a sum of Matsubara frequencies, $\zeta_n = 2\pi n/\hbar\beta$ [11],

$$F(L)=\sum_{n=0}^{\infty}{}'\, g(L,i\zeta_n),\quad \beta=\frac{1}{k_BT}, \tag{1}$$

where the spectral function $g(L,i\zeta_n)$ obtains contributions from the transverse electric (TE) and the transverse magnetic (TM) modes,

$$g(L,i\zeta_n)=\frac{1}{\beta}\int\frac{d^2k}{(2\pi)^2}\Big\{\ln\Big[1-e^{-2\gamma_2 L}r_{21}^{\mathrm{TE}}r_{23}^{\mathrm{TE}}\Big] +\ln\Big[1-e^{-2\gamma_2 L}r_{21}^{\mathrm{TM}}r_{23}^{\mathrm{TM}}\Big]\Big\}. \tag{2}$$

Here, $\gamma_i=\sqrt{k^2+(\zeta_n/c)^2\varepsilon_i}$, k is the magnitude of the wave vector parallel to the surface, and the prime on the summation sign indicates that the $n=0$ term should be divided by 2. We have used the notations

$$r_{ij}^{\mathrm{TE}}=\frac{\gamma_i-\gamma_j}{\gamma_i+\gamma_j}\quad\text{and}\quad r_{ij}^{\mathrm{TM}}=\frac{\varepsilon_j\gamma_i-\varepsilon_i\gamma_j}{\varepsilon_j\gamma_i+\varepsilon_i\gamma_j} \tag{3}$$

for the TE and TM mode reflection coefficients.

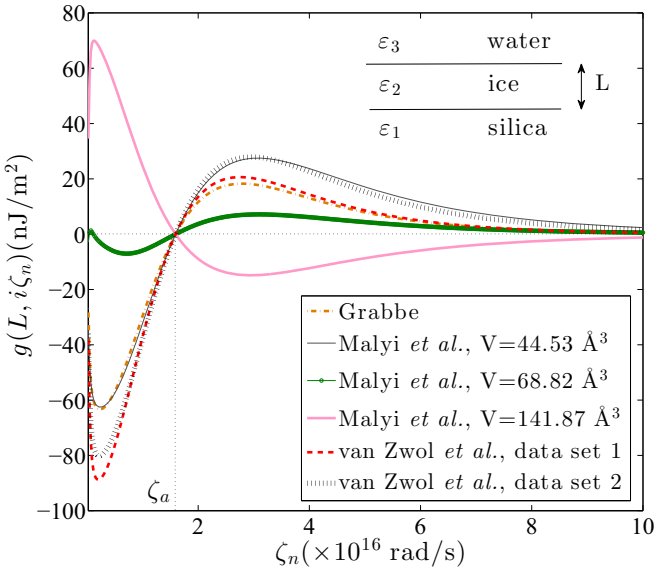

FIG. 3. Spectral function $g(L,i\zeta_n)$ as a function of Matsubara frequency (ζ_n) for a silica-ice-water system with $L = 2.0$ nm thick ice film. We compare the result using different silica dielectric functions presented in Fig. 2. The zero frequency contributions for various silica materials (not shown in the figure) are at least one order of magnitude higher than the contributions from other Matsubara frequencies (≈ -300 nJ/m^2). All the curves vanish at the same point, corresponding to the transition frequency $\zeta_a \approx 1.60 \times 10^{16}$ rad/s.

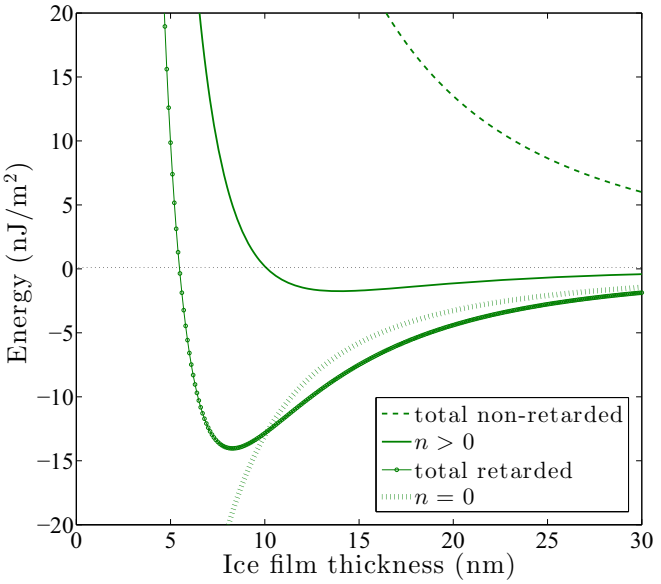

FIG. 4. Contributions to the Lifshitz free energy per unit area for a silica-ice-water ($V = 68.82$ Å^3) system as a function of ice film thickness. Four different curves are shown: the total nonretarded energy, the contributions from the $n > 0$ term to the retarded energy, the total retarded energy, and the contribution from the $n = 0$ term alone.

We use dielectric functions for different silica, each with a specific nanoporosity, or average volume (V) per SiO_2 unit, computed directly from first-principles calculations, as reported in our previous work [24]. However, since the phonon contribution to the dielectric function at imaginary frequencies can have a noticeable impact on the Lifshitz forces, we model the phonon parts of the dielectric functions using the single-phonon Lorentz model and the Kramers-Heisenberg equation [34]. Here, the longitudinal frequency ($\zeta_{LO} = 0.1351$ eV), taken to be the same for all considered systems, is determined from the fitting of the multiphonon contribution to the dielectric function of quartz. The longitudinal and transverse optical frequencies for quartz are taken directly from the experimental data [35]. At the same time, the single-phonon transverse frequency ζ_{TO} is computed from the Lyddane-Sachs-Teller equation [36] using the fitted ζ_{LO} and dielectric constants reported in our previous work [24]. We also use parametrized model dielectric functions for different silica materials based on the optical data and the Kramers-Kronig relationship given by Grabbe [25] and two separate data sets by van Zwol and Palasantzas [26] for comparison. We take the dielectric functions of ice and water at $T = 273.16$ K from Elbaum and Schick [23]. Figure 2 shows the plots of dielectric functions for ice, water, and different silica materials.

In Fig. 3 we plot the spectral function $g(L,i\zeta_n)$ in Eq. (2), for different silica-ice-water systems, at $L = 2.0$ nm. The total Lifshitz energy is the area under the curve(s), getting positive contributions from the positive area and negative contributions from the negative area. The cancellation between these contributions results in a dominant role for the $n = 0$ Matsubara term. In the symmetric systems involving water ($\varepsilon_1 = \varepsilon_3$) the large static dielectric permittivity of water compared to the interacting media causes an increase in the factor $r_{21}^{TM}(0)r_{23}^{TM}(0) \approx 0.9$. This enhances significantly the contribution of the $n = 0$ term to the total interaction energy [15]. By contrast, in our asymmetric silica-ice-water system the above factor is approximately 0.02 due to very similar values of the static dielectric permittivities of ice and water. The contribution of the $n = 0$ term is therefore not enhanced here.

We nevertheless find that the $n = 0$ Matsubara term is crucial for all separation distances, as shown in Fig. 4. It is evident from the plot that if we ignore the retardation effect, then there will be a complete freezing of the water, which, however, is not a natural phenomenon. The contribution to the Lifshitz energy from the $n = 0$ term is always attractive and considerably influences the equilibrium thickness as well as the stability of the ice sheet as compared to the contributions from the $n > 0$. This conclusion is true for most materials with a low dielectric constant that can serve as nucleation sites for ice formation but not for metals, where the $n = 0$ term gives a repulsive contribution.

An estimate for the stable thickness of ice formed at the interface of the silica-water system is obtained [29] by replacing the two exponentials in Eq. (2) with a step function, $e^x \sim \theta(x)$. This corresponds to $2\gamma_2 L \approx 1$, which leads to $L \approx c/2\zeta_a\sqrt{\varepsilon_2(\zeta_a)} = 7.9$ nm. This estimate is similar to the equilibrium thicknesses of the ice sheets for the broad range of the silica-water interfaces calculated using the complete Lifshitz energy of Eq. (1), shown in Table I. This stable thickness corresponds to an extremum in the plots of the total Lifshitz energy versus the separation distance L in Fig. 5. The last column in Table I shows the relative contribution of the $n = 0$ term with respect to the total energy at the equilibrium

TABLE I. Ice film thickness at different silica-water interfaces at the triple point of water. The stable equilibrium ice film thickness is shown in the middle column. A zero value corresponds to the absence of a stable equilibrium at small distances for high nanoporosity silica material. In the last column we show the ratio between the $n = 0$ term and the total retarded Lifshitz energy at the equilibrium ice film thickness. The plots for volumes 35.68 and 106.39 Å^3 are not shown in Figs. 3 and 5.

Volume (Å^3)	Ice film thickness (nm)	$F^{\rm eq}_{n=0}/F^{\rm eq}$
35.68	7.0	0.20
44.53	7.0	0.30
56.96	7.2	0.57
68.82	8.3	1.34
106.39	0	
141.87	0	
Grabbe	2.6	0.59
Data set 1	0.9	1.29
Data set 2	5.4	0.28

thickness. It is clear that the contribution from the $n = 0$ term is dominant in most cases, even at the small separation distances, and even exceeds the contribution coming from the $n > 0$ terms in some cases.

Typically for the Casimir interaction between two atoms, retardation effects become relevant for distance regimes set by the cube root of the polarizability of the atoms, which serves as the scale for the retardation effects. In our system, the characteristic frequency is the transition frequency ζ_a, which sets the scale for retardation to be 8 nm. This includes the speed of light in the intermediate medium.

We summarize our results for ice formation near silica surfaces in Fig. 5 and Table I. We find that the system shows the behavior of the vapor-ice-water interface of Ref. [30], i.e., the intermediate layer vanishes, for very high nanoporosity (large

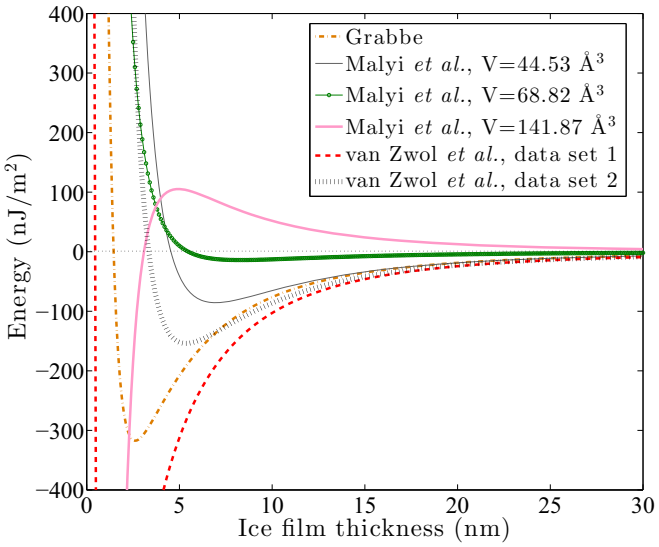

FIG. 5. Lifshitz free energy per unit area for silica-ice-water systems as a function of the ice film thickness using different silica dielectric functions presented in Fig. 2.

V for the SiO_2 material). The spectral function $g(L, i\zeta_n)$ in this case is reversed (see Fig. 3). In this limit when the substrate behaves more as a vapor, there is no ice growth due to Lifshitz forces, as predicted by Elbaum and Schick [30]. For these cases, due to the attractive $n = 0$ contribution, there is a global energy maximum around $L = 4 - 5$ nm and a local very weak energy minimum around $L = 1 - 2\,\mu$m. However, for a large range of different silica materials, we predict a surface specific ice growth near the silica-water interface. The transition point between a stabilized thin ice layer and destabilized ice growth is apparent from the dielectric spectrum of nanoporous silica, seen in Fig. 2. The stable thin layer is lost when the silica porosity is high enough to cause its dielectric function to remain below that of ice and water.

The study of ice formation at a silica interface has significant applied value as the model system for how water freezes on glass, rocks, and soil surfaces. Quasiliquid layers are observed to form on solid-ice interfaces, depending on the surface density and roughness [37–41]. Optical reflection measurements have demonstrated the existence of up to a few tens of nanometer thick premelted water sheets on ice crystal surfaces [42–45]. Ice in contact with silica has been found to have a 5–6 nm thick quasiliquid layer on the surface with a density similar to high-density amorphous ice [37]. Several measurements have been carried out aiming at an understanding of the structure of the ice surface [46–49]. For a thorough review on the premelting of the ice, see Dash *et al.* in Ref. [31]. From our study, we find that the Lifshitz force promotes freezing in the limit of low porosity, analogous to the reduction in the premelting layer observed with decreasing temperature [41]. In another experimental study, Bluhm and Salmeron [50] observe a 0.7 nm thin sheet of ice formed at the mica-water interface. We obtain a thickness 2.7 nm for ice formation on mica using the above techniques with the dielectric permittivity of mica from Ref. [51].

In real systems, optical properties, surface charges, surface roughness [3], the density of the material, gravity [28,52], ions [40,53,54], the presence of gas layers on ice premelting in pores [32], and so on influence the total energy of the system. It is an advantage of the theory that different properties can be analyzed separately.

In summary, the investigations of ice growth, due to the Lifshitz interaction, near different materials require a detailed knowledge of the dielectric functions for a large range of frequencies. The zero frequency term, although of fundamental interest in its own right, can, under specific circumstances, also play a major role in determining the stability and thickness of a thin layer near surfaces at much shorter distances than one would normally expect. Elbaum and Schick observed that the Lifshitz interaction is not sufficient, by itself, to promote ice growth at the water-vapor surface [30]. In contrast, we predict a growth of nanosized ice films driven by the Lifshitz interaction at certain silica interfaces in ice-cold water. We suggest that it should be possible to measure them, perhaps with the use of already available experimental techniques [55].

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