Surface instability and isotopic impurities in quantum solids

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In this paper we employ a self-consistent harmonic approximation to investigate surface melting and local melting close to quantum impurities in quantum solids. We show that surface melting can occur at temperatures much lower than the critical temperature $T_c$ of the solid phase instability in the bulk. Similar effects are driven by the presence of an isotope substitution. In this latter case, we show that stronger local lattice fluctuations, induced by a lighter isotope atom, can induce local melting of the host bulk phase. Experimental consequences and the possible relevance in solid helium are discussed.

I. INTRODUCTION

Although melting is a very common phenomenon in nature, the debate about its microscopic mechanism is still open.\textsuperscript{1-3} The first empirical theory was advanced by Lindemann.\textsuperscript{4} According to this view, melting occurs when the ratio between the root mean square (rms) $v_{\text{rms}} = \sqrt{\langle u^2 \rangle}$ of the thermally activated lattice fluctuations and the lattice constant $a$ exceeds a phenomenological threshold $v_{\text{rms}}/a \gtrsim 0.16$ which is roughly material independent.\textsuperscript{5-6} In spite of its several flaws (melting is described in terms of the properties of only the solid phase; no cooperative process and no role of defects are considered...), this simple criterion seems to work reasonably well for a variety of materials.\textsuperscript{7} Lindemann criterion has been recently found to apply as well at a local level around crystal defects.\textsuperscript{8,9} This large range of validity of the Lindemann criterion suggests thus that a microscopic mechanism is actually operative.

The most simple (and employed) model to account for the Lindemann phenomenology is the self-consistent harmonic approximation (SCHA). This maps an anharmonic phonon model in a harmonic one. Anharmonicity is, then, taken into account, at a mean-field level, through a Debye-Waller-like term which is evaluated self-consistently. The breakdown of this approach is interpreted as a signal of solid phase instability, and hence related to melting. One of the strength of this theory is that it predicts, in contrast with the Born criterion but in agreement with the experimental observation, a partial but not total softening of the elastic constants of the bulk.

The SCHA represents moreover an efficient tool to understand in a qualitative way the phenomenon of the surface melting (SM), as first proposed by Pietronero and Tosatti (PT).\textsuperscript{10} In this context, the physical mechanism underlying the surface melting is quite simple: atoms close to the surface have larger lattice fluctuations due to the reduced number of nearest neighbor sites, and the SCHA breaks down consequently at smaller temperatures than in the bulk. It is clear that this simple theory does not represent an exhaustive description of the surface melting phenomenology, which should include roughening, preroughening, partial wetting, the role of “crystallinity” etc.\textsuperscript{11-12} In addition it should be stressed that the SCHA does not determine directly the melting point but rather the instability of the solid phase which is prevented by the melting process itself.\textsuperscript{13} In this perspective this criterion should not be employed at a quantitative level. Nevertheless, since the solid phase instability and the actual melting process are usually related to each other, the PT theory provides a simple and useful way to get information about the tendency of a system towards melting and surface melting and their dependence on microscopic parameters.

In this paper we generalize the results of the PT approach in the case of quantum solids. The Lindemann criterion in the quantum solid is shown to be twice as large as the one in the classical limit, in agreement with experimental reports.\textsuperscript{14} We show a phase diagram for both the bulk and surface melting cases and we investigate also the local melting due to an isotopic substitution. The temperature dependence of the lattice fluctuations for the different classical/quantum regimes is evaluated and also the profile of the lattice fluctuations as function of the distance from the surface or the isotopic impurities. The paper is organized as follows: in Sect. III we review the approach of Pietronero-Tosatti for classical solids; in III we generalize the PT approach to investigate bulk properties in quantum solids; surface melting and solid phase instability close to a quantum isotope impurity are analyzed respectively in Sect. IV and Sect. V. Finally, in Sect. VI we discuss our results and draw some final conclusions.

II. SCHA AND SOLID PHASE INSTABILITY IN BULK AND ON SURFACES

Let us consider for simplicity a one-dimensional chain of atoms. At the harmonic level, we can write the equations of motion for the lattice displacement $u_n$:

\begin{equation}
M \ddot{u}_n + \frac{k_{n,n+1}}{2} (u_n - u_{n+1}) + \frac{k_{n,n-1}}{2} (u_n - u_{n-1}) = 0, (1)
\end{equation}
where $M$ is the atomic mass, $n$ denotes the site index. The constant forces $k_{n,n'}$, at the harmonic level, are related to the inter-atom potential $V_{n,n'}$ through the relation $k_{n,n'} = \partial^2 V_{n,n'}/\partial u_n^2$. Writing the potential $V_{n,n'}$ in terms of a Fourier expansion, $V_{n,n'} = \sum_q V_q \exp[iq(u_n - u_{n'})]$, we have thus, at the harmonic level, $k_{n,n'} = k_0 = -\sum_q q^2 V_q$.

In the spirit of SCHA, anharmonic terms can be taken into account, by replacing the constant forces $k_{n,n'}$, evaluated at the lattice equilibrium, with their expectation value $\tilde{k}_{n,n'}$ averaged over the lattice fluctuations. We have thus explicitly:

$$\tilde{k}_{n,n'} = \left\langle \frac{\partial^2 V_{n,n'}}{\partial u_n^2} \right\rangle$$
$$= -\sum_q q^2 V_q \exp[-q^2 \left\langle |u_n - u_{n'}|^2 \right\rangle / 2]$$
$$\simeq k_0 \exp[-\lambda \left\langle u_n^2 \right\rangle / 2 - \lambda \left\langle u_{n'}^2 \right\rangle / 2], \quad (2)$$

where in the last line we have neglected the cross terms and we have replaced the dependence on the momenta in the exponential with an effective parameter $\lambda$.

By inserting (2) in Eq. (1) and considering the motion of each atom as an Einstein oscillator we have:

$$M \ddot{u}_n + \left[ \tilde{k}_{n,n+1} + \tilde{k}_{n,n-1} \right] u_n = 0, \quad (3)$$

where anharmonic effects are taken into account in the self-consistent renormalization of the elastic constants $\tilde{k}_{n,n'}$. Note that $\tilde{k}_{n,n'}$ depends on the expectation value of the quadratic lattice fluctuations on both sites $n, n'$. It follows that the atomic motion described in Eq. (3) is ruled by the lattice fluctuations of the lattice environment. In a bulk system $\left\langle u_n^2 \right\rangle = \left\langle u_n^2 \right\rangle = \left\langle u_n^2 \right\rangle$, then

$$\tilde{k}_{n,n'} = \tilde{k} = k_0 \exp\left[-\lambda \left\langle u_n^2 \right\rangle \right],$$

and we get an unique self-consistent equation

$$\left\langle u_n^2 \right\rangle = \frac{k_B T}{\tilde{k}} = \frac{k_B T}{k_0} \exp[\lambda \left\langle u_n^2 \right\rangle], \quad (5)$$

where $k_B$ is the Boltzmann constant. In similar way, the SCHA phonon frequency is given by $\tilde{\omega}_0 = \sqrt{\tilde{k}/M} = \omega_0 \exp[-\lambda \left\langle u_n^2 \right\rangle / 2]$, where $\omega_0 = \sqrt{k_0/M}$ is the bare phonon frequency at the purely harmonic level. It is convenient to rewrite Eq. (3) by introducing the dimensionless quantities $y = \lambda \left\langle u_n^2 \right\rangle$, $\tau_{cl} = k_B T/M$: $y(\tau_{cl}) = \tau_{cl} e^{y(\tau_{cl})}$, $\psi(\tau_{cl}) = y(\tau_{cl})$, $\psi_{cl} = k_B T / M$: $y(\tau_{cl}) = \tau_{cl} e^{y(\tau_{cl})}$, $\psi(\tau_{cl}) = y(\tau_{cl})$. Eq. (3) has no solution for $\tau_{cl} \geq \tau_{cl}^{max} = 1/e = 0.368$, which determines a critical temperature $k_B T_c = 0.368 k_0 / \lambda$. At this value $y(\tau_{cl}^{max}) = 1$ and the maximum magnitude of the allowed lattice fluctuations above which the solid phase is unstable is $\langle u_n^2 \rangle^{max} = 1/\lambda$. Note that $\langle u_n^2 \rangle^{max}$ depends neither on the atomic mass nor on the force constant $k_0$, in agreement with the observation of a material independent Lindemann criterion.

Eq. (3) represents also the starting point to apply the SCHA to surface melting. In this case, one defines a local average lattice fluctuation $\langle u_n^2 \rangle$ which depends on the site index $n$. In the same spirit one can define a local elastic constant:

$$\tilde{k}_{n,n-1,n+1} = \left[ \tilde{k}_{n,n+1} + \tilde{k}_{n,n-1} \right] = k_0 e^{-\lambda \left\langle u_n^2 \right\rangle / 2} \left[ e^{-\left\langle u_{n-1}^2 \right\rangle / 2} + e^{-\left\langle u_{n+1}^2 \right\rangle / 2} \right] \quad (7)$$

We can write thus a set of recursive equations where the lattice fluctuations of the atom $n$ depend on the lattice fluctuations of the $n-1$ and $n+1$ atoms. The recursion is truncated at the atom $n = 1$ which represents the outer atom close to the free surface. This atom probes an effective harmonic potential smaller than the bulk, which increases its tendency towards melting. A numerical solution shows that the solid phase for the surface atoms becomes unstable at $\tau_{cl}^{SM} = 0.271$, $26 \%$ smaller than the bulk value. The same theory permits to evaluate the profile of the lattice fluctuations as function of the distance from the surface. These theoretical predictions agree quite well with the profile of the lattice fluctuations close to defects (grain boundaries, dislocations, vacancies) in colloidal solids. Note that, although the temperature of surface melting is smaller than in the bulk, local lattice fluctuations of the outer atoms can be larger than the ones in the bulk, violating locally the Lindemann criterion. This is also in agreement with Ref. [3]. For instance, for the outer atoms $n = 1$ one finds $y(\tau_{cl}^{SM}) = 1.74$. This is $74 \%$ larger than the value in the bulk.

### III. BULK PROPERTIES OF QUANTUM SOLIDS

We generalize now the above theory to the case of quantum solids. In the following we shall assume a one-particle picture to be still valid, because of the smallness of the exchange terms in the solid phase ($J^{max} \sim 0.1 K$ in $^4$He, $J^{max} \sim \mu K$ in $^3$He) with respect to the melting temperatures $T_m \geq 2 K$ [15,16]. On the other hand a major role in our approach will be played by the quantum fluctuations which dominate at low temperature in the quantum regime. According to this perspective, the atomic motion of the atom $n$ is described in terms of the SCHA Hamiltonian of the quantum oscillator:

$$\left[ -\frac{\hbar^2 \nabla_n^2}{2M} + \frac{1}{4} k_{n,n-1,n+1} u_n^2 \right] \psi(u_n) = E \psi(u_n), \quad (8)$$

where the self-consistent expression for the local potential $\tilde{k}_{n,n-1,n+1}$ is reported in Eq. (7).

We consider first the melting properties of bulk systems ($\tilde{k}_{n,n-1,n+1} = 2k$). In this SCHA quantum model
the total amount of lattice fluctuations is now easily computed as:

$$\langle u^2 \rangle = \frac{\hbar}{2M\omega_0} \left[ 1 + 2n\left(\frac{\hbar\omega_0}{k_BT}\right) \right], \quad (9)$$

where $n(x) = 1/[e^x - 1]$ is the Bose factor and where we remind $\omega_0 = \sqrt{k/M}$ and $k$ is given by Eq.\[4\]. In the classic limit $k_BT \gg \hbar\omega_0$, $n(x) \simeq 1/x \gg 1$, and we recover the classical result of Eq. (4). On the other hand, in the zero temperature limit, lattice fluctuations are due only to zero point quantum motion. In this case $n(x) = 0$ and Eq. (9) reads:

$$\langle u^2 \rangle = \frac{\hbar}{2\sqrt{MK}} = \frac{\hbar}{2\sqrt{MK}_0} \exp \left[ \lambda\langle u^2 \rangle/2 \right], \quad (10)$$

which, introducing the variable $\tau_Q = \hbar/2\sqrt{k_0M}$, can be written in the dimensionless form:

$$y(\tau_Q) = \tau_Q e^{y(\tau_Q)/2}. \quad (11)$$

Eq. (11) represents the quantum generalization of Eq. (9), where the instability of the solid phase is now triggered by the magnitude of the quantum lattice fluctuations. This occurs for $\tau_Q \geq \tau_Q^{\text{max}} = 2/e = 0.736$. It is interesting to note that the breakdown of the solid phase driven by quantum fluctuations is not merely equivalent to the one related to the thermal motion. Indeed, for a quantum solid, we would predict a maximum magnitude of lattice fluctuations $y(\tau_Q^{\text{max}}) = 2$, two times larger than for classical solids. This behavior is indeed in agreement with the report of the Lindemann ratio $u_{\text{rms}}/a \simeq 0.28$ in helium solids\[11,17,18\] to compare with $u_{\text{rms}}/a \simeq 0.16$ for classical solids.

We also consider now the general case where both thermal and quantum fluctuations are important. From Eq. (9), after few straightforward passages, we get

$$y(\tau_Q, \tau_{cl}) = \tau_Q e^{y(\tau_Q, \tau_{cl})/2} \left[ 1 + 2n\left(\frac{2\tau_Q}{\tau_{cl}} e^{-y(\tau_Q, \tau_{cl})/2} \right) \right]. \quad (12)$$

Eq. (12) generalizes the stability criterion based on the SCHA in the full quantum-thermal case. As a general rule we can expect that the classical regime is relevant in the empirical range $k_BT/\hbar\omega_0 \gtrsim 1/4$, which corresponds to $\tau_Q \lesssim 2\tau_{cl}$, while in the opposite regime $\tau_Q \gtrsim 2\tau_{cl}$ quantum effects are dominant.

In Fig. 1 we show the phase diagram in the full $\tau_Q$-$\tau_{cl}$ space where the instability of the SCHA occurs. Along the boundary line, the critical lattice fluctuations increase smoothly from $y = 1$ in the $\tau_Q = 0$ case to $y = 2$ in the $\tau_{cl} = 0$ case. Also interesting is the dependence of the lattice fluctuations as function of $\tau_{cl}$, namely the temperature (Fig. 1 bottom panel). In the classical case, $\tau_Q = 0$, the quadratic fluctuations $y \propto \langle u^2 \rangle$ increase linearly with $\tau_{cl}$ until anharmonic effects take place. Anharmonicity is reflected in a upturn of the temperature dependence of $y(\tau_{cl})$ and eventually in the breakdown of the solid phase for $\tau_{cl} = 1/e$ and $y = 1$. Increasing $\tau_Q$ leads not only to the presence of zero point motion quantum fluctuations at $\tau_{cl} = 0$, but also to an overall change of the temperature dependence of $y$. In particular, the range of the linear temperature dependence, characteristic of classical harmonic solids, is rapidly reduced and for strongly quantum solids it disappears. Lattice fluctuations are large already at $T = 0$ and they are almost constants in a wide temperature range (note that in this regime anharmonic effects are in any case present due to quantum fluctuations) until an abrupt upturn with the temperature leads to the breakdown of the solid phase. This trends is in good qualitative agreement with recent experimental measurements\[19\] and Quantum Monte Carlo calculations\[20\]. We shall discuss them in details in Sect. VI.
IV. SURFACE MELTING OF QUANTUM SOLIDS

After having investigated the bulk properties of quantum solids, we analyze now the occurrence the role of quantum fluctuations on the surface melting.

We can write a recursive set of equations by considering the quantum/thermal SCHA solution of the $n$-th atom

$$
\langle u_n^2 \rangle = \frac{\hbar}{2M\bar{\omega}_n} \left[ 1 + 2n \left( \frac{\hbar\bar{\omega}_n}{\kappa_B T} \right) \right],
$$

(13)

where $\bar{\omega}_n = \sqrt{\tilde{k}_{n,n-1,n+1}/2M}$ and where the local elastic constant $\tilde{k}_{n,n-1,n+1}$ is still given by Eq. (7). Employing the usual dimensionless variables $\tau_Q, \tau_{cl}, y_n$, we can thus write:

$$
y_n = \frac{\sqrt{2\tau_Q e^{yn/4}}}{e^{-yn/2} + e^{yn/2}} 
\times \left[ 1 + 2n \left( \frac{2\tau_Q e^{yn/2} + e^{-yn/2}}{\sqrt{2}e^{yn/4}} \right) \right]
$$

(14)

which is valid for any $n \geq 2$, while the outer atom $n = 1$ obeys the relation

$$
y_1 = \sqrt{2\tau_Q e^{(y_1+y_2)/4}} \left[ 1 + 2n \left( \frac{2\tau_Q e^{-(y_1+y_2)/4}}{\sqrt{2}\tau_{cl}} \right) \right]
$$

(15)

In order to obtain a numerical solution of Eqs. (14)-(15) for given $\tau_Q, \tau_{cl}$ in the stable solid phase, we start by choosing a trial value of $y_1$. The full set of $\{y_n\}$ is thus obtained by Eqs. (14)-(15). The initial trial value of $y_1$ is thus varied until $y_{n=\infty}$ converges to its bulk value. Typically, this is the only physical solution, since $y_{n=\infty}$ diverges for larger values of $y_1$, while it becomes rapidly negative for smaller values of $y_1$. For $\tau_Q, \tau_{cl}$ larger than some critical value, the procedure does not converge for any value of $y_1$, signifying that the solid phase of the surface atom, described by the SCHA, is unstable.

The resulting phase diagram, in the full $\tau_Q$-$\tau_{cl}$ space, is shown in Fig. 2 (top panel), where we compare the boundary of the surface melting instability (dashed line) with the one of the bulk melting (solid line). For the pure quantum case, $\tau_{cl} = 0$, at zero temperature the surface instability occurs for $\tau_Q^{SM} = 0.664$ where the lattice fluctuations of the outer atoms become as large as $y_{1,SM} = 3.21$. It is interesting to notice that, for $0.664 < \tau_Q < 0.736$, the surface is unstable even at zero temperature whereas the bulk solid phase is always stable up to a finite temperature range. The ratio $T_{SM}^{SM}/T_c$ between the surface melting temperature and the temperature of bulk melting is shown in the bottom panel of Fig. 2, showing that the critical temperature of surface melting can be significantly lower than the bulk one in quantum solids.

Before concluding this section, we would like to briefly compare the melting occurring at a free surface with other cases such as grain boundaries. In the case of a free surface, in going from Eq. (14) to Eq. (15), we have dropped in Eq. (15) the contribution of the $n = 0$ atom. We note that the same results would be obtained in Eq. (15) considering $n = 1$ and assuming the lattice fluctuations at the site $n = 0$ to be infinite, namely $y_{n=0} = \infty$. This latter condition would be obtained by the harmonic oscillator solution of (15) at the site $n = 0$ with a vanishing elastic constant $\tilde{k}_{n,n-1,n+1}$, and it express nothing more than the condition that atoms for $n < 1$ are not in a solid arrangement but in a gaseous phase.

An intermediate situation is encountered when melting at grain boundary interfaces is considered. In this case the outer atom $n = 1$ of a grain would not probe a free surface at the site $n = 0$, but it will interact with a lattice environment with a different arrangement. These two situations can be described by a similar set of recursion relations (14), but with different boundary conditions: in the free surface case boundary conditions at site $n = 0$ will be described by a completely soft oscillator $k_{n,n-1,n+1} = 0$, signalizing that bulk solid is interfaced...
with a free gaseous phase; on the other hand, in the case of grain boundaries, the outer atom \( n = 1 \) will still probe a crystal structure for \( n \leq 0 \), although with a different arrangement. The boundary conditions at site \( n = 0 \) will be still described thus by Eq. (13), but with a not completely soft mode. We expect thus that melting processes occur as well at grain boundaries as in the case of free surface. From the mathematical point of view, this situation is identical to the case of quantum isotopic substitutions, and it will be discussed in details in the next section.

V. QUANTUM MELTING DRIVEN BY ISOTOPIC IMPURITIES

In this section we address the problem of the solid phase stability close to a single local isotopic substitution embedded in a perfect lattice structure. In the SCBA approach, local stability of the solid phase is given by the solution of Eq. (13). It is easy to check that, in the classical limit \( k_B T \gg \hbar \omega_n \), the dependence on the atomic mass \( M \) in Eq. (13) drops out, so that different isotope solids should probe the same stability conditions. On the other hand, the mere observation of a different melting line for \(^4\)He and \(^3\)He is a direct evidence that helium is in a quantum regime.\(^{21,22}\) Different isotopes are thus expected to affect the bulk solid phase stability. We expect the same at the local level.

In the following we shall consider the case of a isolate substitution with a lighter isotope in a host matrix of heavier atoms. Quantum fluctuations in the two cases will be ruled locally by the parameters \( \tau_L = \lambda L / 2 \sqrt{k_B M_L} \), \( \tau_H = \lambda H / 2 \sqrt{k_B M_H} \), respectively for the lighter (L) and for the heavier (H) atoms. To study the stability of the solid phase close to this isotopic quantum impurity, we can still employ the recursive relations (14), namely for \( n \leq -1 \), \( n \geq 1 \) we set \( \tau_Q = \tau_H \), whereas for \( n = 0 \) (quantum isotope impurity) we have \( \tau_Q = \tau_L \). We shall consider the representative case of a \(^3\)He impurity embedded in \(^4\)He solid. In this case \( \tau_L / \tau_H = \sqrt{4/3} \).

In Fig. 3 (top panel) we show the phase diagram of the lattice instability of the host \(^4\)He solid close to the quantum isotopic \(^3\)He impurity. It is instructive to compare the classical limit \( \tau_Q = 0 \) with the pure quantum one \( \tau_{cl} = 0 \). In the first case lattice fluctuations of the guest atom, as well as of the host atoms, are independent on the relative atomic mass and they depend only on the temperature. As a consequence, the solid phase close to the guest atom is completely unaffected by the isotopic substitution. A quite different situation occurs in the highly quantum regime \( \tau_{cl} = 0 \). In this case local quantum lattice fluctuations of the lighter guest atom can be significantly enhanced due to its reduced atomic mass, and they can be sufficiently large to induce a local melting of the host solid phase. At \( \tau_{cl} = 0 \) this occurs for \( \tau_H > 0.681 \), not much higher than in the case of a free surface truncation (\( \tau_H > 0.664 \)). Note that Fig. 3 defines a region (quantum impurity melting) where solid phase is still stable in the bulk but local quantum lattice fluctuations break down the solid phase close to the isotopic substitution. On a real physical ground we can expect liquid bubbles of host atoms to appear close to the guest isotope. Unfortunately, since the present analysis is only related to the stability condition of the solid phase, we are not able to estimate the size of the liquid bubble, and more sophisticated approaches are needed. It is interesting to note that, for quantum solids, the critical temperature \( T_{c}^{Q_{imp}} \) for the local stability of the solid phase close to the quantum isotope impurity is reduced with respect to the bulk \( T_{c} \). This is shown in the bottom panel of Fig. 3 where the ratio between the local \( T_{c} \) close to the impurity and the bulk \( T_{c} \) is plotted as function of the quantum degree of the system, parametrized by \( \tau_Q \).
In the quantum regime, where $T^{\text{Qimp}} \lesssim \hbar \tilde{\omega}_0/4$, the local melting temperature $T^{\text{Qimp}}$ can be significantly lower than the one in the bulk $T_c$, and, for $1.39 < \tau_{\text{He}} < 2$, we expect a quantum isotopic impurity to induce local melting down to $T = 0$, although the bulk phase is still stable.

VI. DISCUSSION AND CONCLUSIONS

In this paper we have investigated the stability of quantum solids with respect to surface melting and to isotopic quantum substitutions. Both these phenomena can be essentially related to the amount of lattice fluctuations, and they can be driven thus by thermal fluctuations as well as by the zero point quantum motion. We have shown that the effects of isotopic impurities and surface melting are strongly enhanced in quantum solids. In particular we show that when quantum fluctuations are dominant in quantum solids the solid phase can be rapidly destroyed on the surface and close to quantum impurities at temperatures much smaller than for the bulk melting.

Helium solids are the natural candidates where the quantum instabilities of surface or interface can occur. The actual relevance of these quantum melting effects are of course ruled by the magnitude of the quantum lattice fluctuations which are parametrized in our model by the quantity $\tau_Q$. An accurate calculation of the quantum lattice fluctuations as a function of the temperature in $^4$He and $^3$He solids has been provided recently, by using of Quantum Monte Carlo (QMC) techniques, by Draeger and Ceperley in Ref. 20, in excellent agreement with the experimental data. Quite interestingly, they find that the mean square lattice displacement $\langle u^2 \rangle_T$ does not follow at low temperature an harmonic behavior $\langle u^2 \rangle_T \simeq \langle u^2 \rangle_{T=0} + \alpha T^2$, but rather a more shallow one $\langle u^2 \rangle_T \simeq \langle u^2 \rangle_{T=0} + \beta T^3$.

Ref. 20 represents a suitable source to estimate an effective value of $\tau_Q$ representative of solid helium. To this aim we fit the temperature dependence of the QMC data of Ref. 20 with our quantum SCHA model described by Eq. (12), where only two independent fitting parameters appear, namely $\lambda$ and $k_0$ (remind that $\tau_{cl} = \lambda k_0 T/k_0$, $\tau_Q = \lambda h/2 \sqrt{k_0 M}$). The fit of our quantum SCHA [Eq. (12)] compared with the QMC data is shown in Fig. 4 for three representative cases where the number of numerical data is larger than the number of independent fitting parameters to guarantee the significance of the fitting procedure. Also shown is the fit with a purely harmonic model obtained by setting $\lambda = 0$. The extracted values of $\lambda$ and $k_0$, as well as of the corresponding $\tau_Q$ and of the anharmonic renormalized phonon frequency at $T = 0 \tilde{\omega}_0$ are reported in Table I, where also we report the critical temperature $T_c$ for the solid phase bulk instability evaluated within the SCHA and the experimental melting temperature $T^{\text{m exp}}_{\text{He}}$.

It is worth to comment about the temperature behavior of the QMC data compared with the harmonic ($\lambda = 0$) and anharmonic SCHA fit. An important point to be here underlined is that QMC results show a large mean square lattice displacement at zero temperature all together with a rapidly turn up of $\langle u^2 \rangle$ close to the solid bulk instability. As we have discussed in Sect. III, this is a characteristic trend of highly quantum solids. On the other hand, this behavior is poorly reproduced by a purely harmonic model where the amount of the lat-

| $V_0$ (cm$^3$/mole) | $^{4}$He | $^{3}$He | $^{3}$He |
|-------------------|-------|-------|-------|
| 12.12             | 10.98 | 11.54 |

| $k_0$ (meV/Å$^2$) | $^{4}$He | $^{3}$He | $^{3}$He |
|------------------|-------|-------|-------|
| 110 ± 10         | 140 ± 10 | 150 ± 10 |

| $\lambda$ (Å$^{-2}$) | $^{4}$He | $^{3}$He | $^{3}$He |
|---------------------|-------|-------|-------|
| 14 ± 1              | 15.2 ± 0.8 | 14.7 ± 0.7 |

| $\tau_Q$ | $^{4}$He | $^{3}$He | $^{3}$He |
|----------|-------|-------|-------|
| 0.69 ± 0.08 | 0.66 ± 0.06 | 0.70 ± 0.06 |

| $\tilde{\omega}_0$ (meV) | $^{4}$He | $^{3}$He | $^{3}$He |
|--------------------------|-------|-------|-------|
| 4.6 ± 0.1                | 5.4 ± 0.4 | 6.1 ± 0.5 |

| $T_c$ (K) | $^{4}$He | $^{3}$He | $^{3}$He |
|-----------|-------|-------|-------|
| 14 ± 4    | 20 ± 4 | 18 ± 4 |

| $T^{\text{m exp}}_{\text{He}}$ (K) | $^{4}$He | $^{3}$He | $^{3}$He |
|-------------------------------|-------|-------|-------|
| ~ 15                         | ~ 21  | ~ 22  |

FIG. 4: Lattice fluctuations $\langle u^2 \rangle$ evaluated within the SCHA (solid lines) as function of temperature for different helium solid conditions compared with Quantum Monte Carlo data of Ref. 20. Values of $k_0$ and $\lambda$ in SCHA obtained by fitting QMC data are reported in Table I. Also shown are the purely harmonic fitting of the QMC data with an Einstein and a Debye model.

TABLE I: Values of $k_0$ and $\lambda$ in SCHA obtained by fitting the QMC data of Ref. 20 for three representative helium solids, namely: hcp $^4$He at molar volume $V_0 = 12.12$ cm$^3$/mole, fcc $^4$He at molar volume $V_0 = 10.98$ cm$^3$/mole, and fcc $^3$He at molar volume $V_0 = 11.54$ cm$^3$/mole. Also reported are the corresponding values of $\tau_Q$, the renormalized phonon frequency $\tilde{\omega}_0$ and the predicted critical temperature $T_c$ of the solid phase bulk instability compared with the experimental melting temperature $T^{\text{m exp}}_{\text{He}}$. 

References : 20, 23
tice fluctuations at $T = 0$ is inversely proportional to the temperature dependence. This is even more true if a Debye model would be employed since the temperature dependence of a Debye model is even more shallow than in the Einstein case.

The strong quantum degree of solid helium, qualitatively predicted by these arguments, is confirmed by the numerical analysis of the SCHA fit which predicts a quantum parameter $\tau_Q$ in the range $\tau_Q \approx 0.66 - 0.7$ for the three samples here considered. The robustness of our fits is confirmed by the nice agreement between the critical temperature for the bulk instability of the solid phase estimated by the SCHA and the experimental melting temperature.

These results have important consequences with respect to the surface/grain-boundary melting instability and local melting induced by quantum impurities. The values of $\tau_Q \approx 0.69$, for the low pressure/high molar volume $V_0 = 12.12 \text{ cm}^3/\text{mole}$, is safely larger than the value $\tau_Q^{\text{SM}} \approx 0.664$ where surface melting occurs at zero temperature, and also or the same order and slightly larger even than $\tau_Q^{\text{SM}} \approx 0.681$ where isotopic impurity induced melting also occurs at zero temperature. Although these estimates have to be meant only indicative of the quantum degree of helium solid, they clearly point out that quantum anharmonic effects are large enough in solid helium, for these or larger molar volumes, to enforce surface melting and local melting close to quantum impurities down to zero temperature. Quantum Monte Carlo simulations have actually confirmed premelting at surface between helium solid and Vycor wall and internal interfaces of a pure helium system, although not all possible interfaces undergoes a solid/liquid transition.

These results shed an interesting light also on the recent report of the Non-Classical Rotational Inertia (NCRI) observed in $^4\text{He}$\textsuperscript{28,29}. While it was initially claimed to be an evidence of a supersolid (SS) phase, subsequent experiments showed a strong dependence of the NCRI on the annealing process on the presence of grain boundaries on the amount of $^3\text{He}$ concentration as well as on the freezing procedure. These observations give rise to an alternative hypothesis to the SS phase, namely, that a liquid phase is confined at the grain boundaries and that mass flow is related to superfluidity of the liquid component. Our results confirm this scenario and shed new perspectives about the role of disorder/grain boundaries in solid helium. In particular we provide a natural explanation for the existence of a liquid (and thus probably superfluid) phase at the grain boundaries and we predict a local liquid phase also around $^3\text{He}$ impurities. Local melting close to isotopic $^3\text{He}$ impurities should be thus explicitly considered.

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