

Quantum effects in condensed matter at high pressure

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Abstract. Experimental data characterizing influence of quantum effects on the equation of state and melting of substances at high pressure are reviewed. It is shown that quantum isotope effects tend to increase upon compression of substances with a predominately Coulomb interaction, whereas compression of the ‘van der Waals substances’ reveals the opposite trend. The ‘cold’ melting of ‘Coulomb substances’ at high pressure is discussed.

1. Introduction

As is well known, many-particle systems may behave either as classical or quantum depending on the conditions. In particular, changing the density (pressure) in a system can significantly affect its properties. But, except for phenomena like superconductivity and superfluidity, many-particle quantum effects are always fairly difficult to observe. It turns out, however, that considerable information on the quantum revelations in the natural world may be gained by studying the isotope effect or the mass dependence on properties of the substances. In what follows the experimental data available on isotope effects at high pressures are analyzed. Since all the experimental data touched upon in this paper are for temperatures far below the corresponding Debye temperatures, only the ground state of the substances under study will be considered.

2. A bit of history

According to quantum theory, the ground-state energy of any bound or condensed system of particles contains a dynamic

part, often called the ‘zero-point’ energy. The existence of this energy follows from the Heisenberg uncertainty principle

$$\Delta x \Delta p \geq \hbar, \quad (1)$$

which expresses the relation between the coordinate uncertainty Δx and the momentum uncertainty Δp . From Eqn (1) it follows that localizing a particle unavoidably increases its kinetic energy. Therefore, the zero-point energy necessarily increases as a material is compressed, however, its influence on material properties also hinges on other contributions to the total energy. In the general case, the zero-point energy is a decreasing function of the mass of the comprising particles, but its exact functional form depends on how the particles interact and the phase state of the substance.¹

Clearly, the zero-point energy is especially important for systems made of low-mass weakly interacting particles.² It is for this reason that the helium isotopes ³He and ⁴He — unlike the heavier noble gases — do not crystallize down to the lowest temperatures at low pressures. The zero-point energy contributes significantly to the equations of state of helium, hydrogen, and other light substances and seems to considerably affect processes in the depths of ‘cold’ stars and massive planets.

One of the most interesting quantum effects is the so-called ‘cold’ melting of strongly compressed materials — a phenomenon first considered by Kirzhnits [2, 3]. Somewhat later, this problem was addressed by Abrikosov [4], whose work was followed by a whole series of papers on the subject (see works [5, 6] for a bibliography). Note that in the absence of a theory of melting, works [2, 4] and many

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¹ The Heisenberg uncertainty principle (1) suggests for the kinetic energy the expression $E_k \cong \hbar^2/2m(\Delta x)^2$. Accordingly, for a system of non-interacting particles $E_k \cong \hbar^2/2mV^{2/3}$. For a ‘van der Waals gas’, we have $E_k = \hbar^2/2m(V-b)^{2/3}$. The kinetic part of the zero-point energy of a ‘Debye crystal’ is $E_k = (9/16)(k/m)^{1/2} = (9/16)\Theta_D$. The total zero-point energy in this case equals $E_z = (9/8)\Theta_D$ [1]. Here, m is the mass, V is the volume, the constant b characterizes the ‘inaccessible’ volume, and Θ_D denotes the Debye temperature.

² For a system with the interaction length σ and interaction energy ε , quantum effects are conveniently described by the so-called de Boer parameter $A^* = \hbar/\sigma\sqrt{m\varepsilon}$ [1].

subsequent papers actually employed the Lindemann criterion [7, 8] in their analyses. Earlier, Pomeranchuk [9] had applied a similar approach in an attempt to explain the instability of the helium crystalline phase at atmospheric pressure. Recall that, according to F Lindemann, a crystal melts when the ratio of the mean-square amplitude of atomic vibrations $\langle \delta r^2 \rangle^{1/2}$ to the interatomic distance r reaches a certain critical value, viz.

$$\frac{\langle \delta r^2 \rangle^{1/2}}{r} \cong C. \quad (2)$$

It is easily shown that if particles in the system interact according to the law $\Phi(r) \propto 1/r^n$, then for the zero-point vibrations we have [2–4, 9]

$$\frac{\langle \delta r^2 \rangle^{1/2}}{r} \propto r^{(n/2)-1}. \quad (3)$$

From Eqn (3) it follows that for $n > 2$ the relative amplitude of zero-point vibrations increases with increasing volume.³ On the contrary, in the Coulomb case — or more precisely for a quantum system of point-like charges immersed in the compensating sea — the relative zero-point vibration amplitude [2]

$$\frac{\langle \delta r^2 \rangle^{1/2}}{r_S} \propto r_S^{-1/2} \quad (4)$$

increases as the volume decreases. Here r_S is determined by the condition $V/N = (4/3)\pi r_S^3$.

Thus, according to the picture developed in Refs [2, 4, 9], quantum effects give rise to two types of global phase diagram as shown in Fig. 1. We emphasize, however, that the Lindemann relation (2) does not provide a melting criterion but is in fact a similarity relation valid only for systems of classical particles interacting according to a power law [8].

Nevertheless, noting that in both of the above cases the ratio of the zero-point energy E_z to the static energy E_{st} is proportional to the relative zero-point vibration amplitude:

$$\frac{E_z}{E_{st}} \propto \frac{\langle \delta r^2 \rangle^{1/2}}{r}, \quad (5)$$

relationships (3) and (4) imply that as pressure increases, the importance of the zero-point energy can either increase or

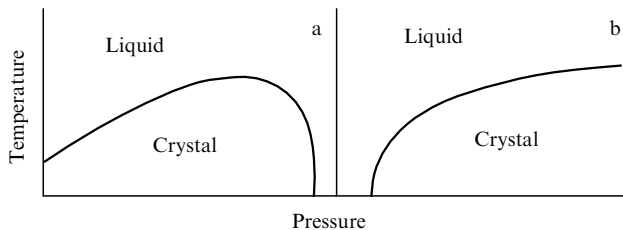


Figure 1. Influence of quantum effects on the melting curve: (a) Coulomb interaction; (b) short-range repulsion.

³ Since for $n < 3$ the energy of a system with a potential of the form $\Phi(r) \propto 1/r^n$ diverges, it is the values $n > 3$ which are physically meaningful.

decrease depending on whether the system has a Coulomb or a short-range power-law interaction.

However, interactions in real materials cannot always be described by $\Phi(r) \propto 1/r^n$, and the pressure dependence of quantum effects remains an open question in the general case. In this connection, undoubtedly the experimental study of quantum effects at high pressures is of interest. Unfortunately, no direct methods are available for measuring the zero-point energy, but, as mentioned earlier, the study of various types of isotope effects provides some insight into the dependence of the zero-point energy on compression.

Most of the experimental data available for the investigation of isotope effects in condensed matter at high pressures have been obtained by studying the equations of state or phase diagrams of isotopically different materials. A standard set of data includes the volume of the material V , pressure P , and temperature T . Combining these with the results of thermodynamic measurements, various macroscopic quantities, including the total energy, can be calculated.

3. Simple estimates

Before discussing experimental data, it will be helpful to estimate the size of macroscopic effects due to the existence of the zero-point energy (see also Ref. [10]).

We start by writing the total energy E_t and pressure P of an arbitrary system at $T = 0$ in the form

$$E_t = E_{st} + E_z, \quad (6)$$

$$P = P_{st} + P_z. \quad (7)$$

The ‘quantum’ pressure P_z is of the same order of magnitude as the zero-point energy density E_z , viz.

$$P_z \propto \frac{E_z}{V}. \quad (8)$$

If quantum effects are small⁴ then, using the trivial relation $P_z = K(\Delta V/V)$, we obtain

$$\frac{\Delta V}{V} \propto \frac{E_z}{KV}. \quad (9)$$

In the Grüneisen quasi-harmonic approximation, Eqns (8) and (9) can be put into a quantitative form by transforming them into Eqns (10) and (12). It is also a simple matter to write a corresponding expression for the bulk modulus, Eqn (11). Thus we have

$$\frac{P_z}{K_0} = \gamma \frac{E_z}{K_0 V_0} \cong \gamma \frac{E_z}{KV}, \quad (10)$$

$$\frac{K_z}{K_0} = \gamma(\gamma + 1) \frac{E_z}{K_0 V_0}, \quad (11)$$

⁴ It is of interest to see which of the macroscopic parameters can serve best as a measure of quantum effects. The answer, it seems, might be the ratio E_z/E_{st} . However, unlike the model systems mentioned in the text, the static energy E_{st} in a real system alternates in sign, making the ratio meaningless for moderate compressions. On the other hand, the available macroscopic variables suggest two combinations with the dimensions of energy: PV and KV (P is the pressure, K the bulk modulus, V the volume). It is readily seen that the product KV is the only scale factor possible here. Notice that for stable states, the quantity KV is always positive and is approximately proportional to the total energy at high pressures.

$$\frac{\Delta V}{V_0} \cong \frac{P_z}{K_0} = \gamma \frac{E_z}{K_0 V_0} \cong \gamma \frac{E_z}{KV}. \quad (12)$$

Here it is assumed that the Grüneisen constant γ is independent of the volume. The index '0' is used to indicate the 'classical' value of the physical quantity so labelled. Since these values are generally unknown, the current values of the corresponding quantities will be used throughout. This does not present any problems if quantum effects are small. Otherwise, the truth of relations (10)–(12) is itself in doubt.

Attention is drawn to the fact that it is the parameter E_z/KV which actually determines the relative magnitude of macroscopic quantum effects in a condensed system.

Relations (10)–(12) are difficult to apply because quantum contributions to thermodynamic quantities cannot be measured directly. Differential effects, however, can be measured and, as mentioned earlier, it is these measurements which allow one to draw some conclusions about the nature and evolution of quantum contributions to the macroscopic properties of the system. We wish to emphasize that a differential effect here means the difference in the observed properties of two isotopically different substances.⁵

From the relations (10)–(12), the following expressions are derived for differential quantum effects:

$$\frac{\Delta P^{mn}}{K} \cong \gamma \frac{\Delta E_z}{KV}, \quad (13)$$

$$\frac{\Delta K^{mn}}{K} \cong \gamma(\gamma + 1) \frac{\Delta E_z}{V}, \quad (14)$$

$$\frac{\Delta V^{mn}}{V} \cong \gamma \frac{\Delta E_z}{KV}. \quad (15)$$

The indices m and n in Eqns (13)–(15) denote the mass numbers. Expressions (13)–(15) are entirely suitable for carrying out concrete estimates,⁶ for which purpose the approximation $\Delta E_z \cong \Delta \Theta_D$ can be employed.

The relations above suggest several equivalent ways of presenting experimental data when analyzing isotope effects. The quantity $\Delta V^{mn}/V$ may prove preferable for study because calculating it from P – V data is a simple procedure introducing virtually no additional errors into the analysis.

We proceed now to discuss the available experimental data. To understand the general situation, it is worthwhile to first discern Fig. 2, in which the pressure dependence of contributions to the total energy is shown for the case of ^4He . It can be seen that in the present case the zero-point energy E_z increases somewhat more slowly with pressure than does the total energy E_t .

⁵ Note that H London's [11] differential equation for the bulk isotope effect,

$$\frac{m}{V} \frac{dV}{dm} = -\frac{9}{16} \gamma \frac{\Theta_D}{KV} \quad (m \text{ is the mass number}),$$

is obtained simply by differentiating Eqn (12).

⁶ As an example, let us try to estimate the bulk isotope effect for lithium isotopes [12]. Taking into account that natural lithium principally consists of the isotope ^7Li and assuming that $\Theta_D^{7\text{Li}} = 344$ K, we have $\Theta_D^{6\text{Li}} = 372$ K. Taking now $V = 12.7$ cm³ mol⁻¹ and $K = 12$ GPa, Eqn (15) gives

$$\frac{\Delta V}{V} \approx \frac{V^{6\text{Li}} - V^{7\text{Li}}}{V^{7\text{Li}}} = 1.5 \times 10^{-3}.$$

Curiously, much more sophisticated estimates based on phonon spectrum calculations for Li [13] yield $\Delta V/V = 1.8 \times 10^{-3}$, to be compared with the experimental value of about 1.2×10^{-3} at room temperature [14].

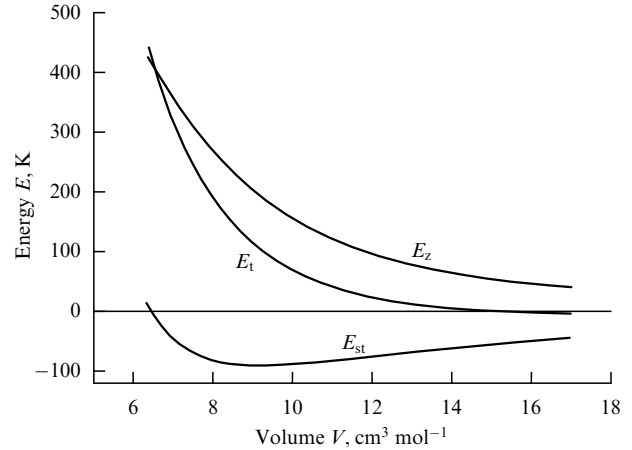


Figure 2. Energy relations for solid ^4He : $E_t = E_{st} + E_z$ is the total energy calculated as $E_t = E_t^0 + \int_{V_0}^V p dV$ ($E_t^0 = -1.35$ K, $V_0 = 17$ cm³ mol⁻¹ [15], from the P – V data of Ref. [16]); the static energy E_{st} was estimated from the expression $E_{st} = 2\varepsilon[12.13229(\sigma/r)^{12} - 14.45489(\sigma/r)^6]$, where $\varepsilon = 10.22$ K, $\sigma = 2.556$ Å.

4. Review of experimental data

4.1 Equations of state

Helium. From Fig. 2 it can be seen that quantum effects are very strong in condensed helium at moderate pressures. The equilibrium volume of ^4He is approximately twice its classical value. Therefore, the relations obtained under conditions of small quantum effects are generally invalid. It is clear, however, that even in this case dimensionless quantities of the form $\Delta V/V$ and $\Delta P/K$ are adequate in describing quantum effects.

Figure 3 shows the results of P – V measurements for ^3He and ^4He [16]. Also shown in the figure are the calculated

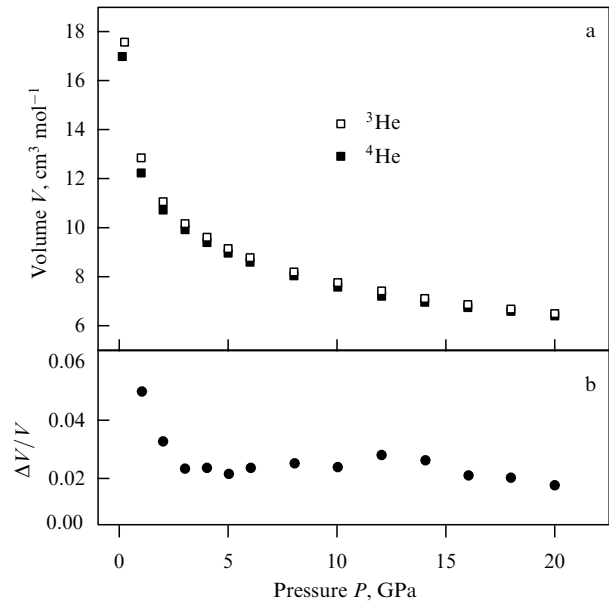


Figure 3. (a) Compression isotherms for ^3He and ^4He [16] at 4 K. (b) Pressure variation of the relative volume difference of ^3He and ^4He , $\Delta V/V = [V(^3\text{He}) - V(^4\text{He})]/V(^4\text{He})$; the data are from Ref. [16].

values of the quantity

$$\frac{\Delta V}{V} \cong \frac{V(^3\text{He}) - V(^4\text{He})}{V(^4\text{He})}.$$

What is surprising here is that the quantity $\Delta V/V$ is relatively constant in the pressure range 5–20 kbar. One should not, however, jump to conclusions about the behavior of $\Delta V/V$ with pressure, because the values of $\Delta V/V$ in this case are comparable to the experimental errors in the volume measurements made in Ref. [16].

Hydrogen and deuterium. Figures 4a and 4b show the compression curves and $\Delta V/V$ values for hydrogen isotopes, based on data obtained in two independent series of experiments [17, 18]. There is a lack of consistency among the experimental data. We note in this connection that the data of Ref. [18] appear to be more trustworthy. Another point to make is that according to Ref. [19], the relative difference in the volume $\Delta V/V$ for hydrogen and deuterium is $\approx 6 \times 10^{-4}$ at a pressure of about 100 GPa.

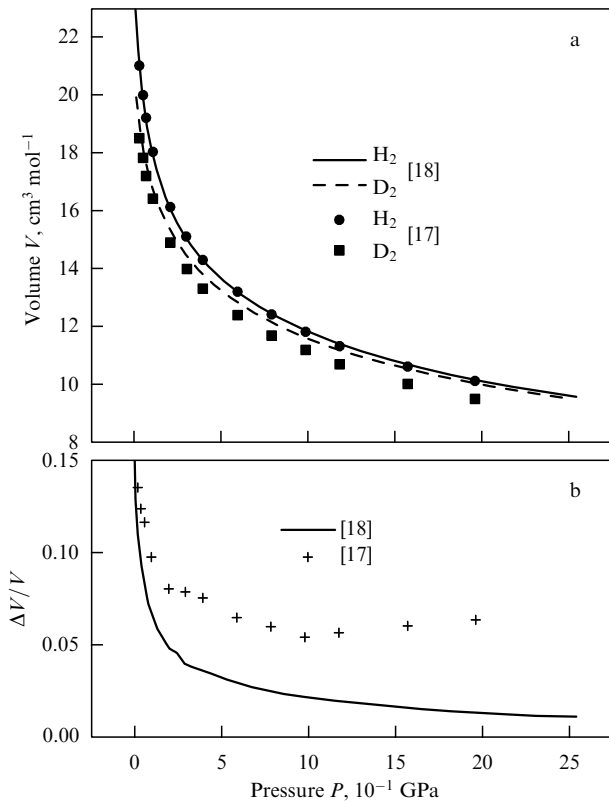


Figure 4. (a) Compression isotherms for H_2 and D_2 at 4 K [17, 18]. (b) Relative volume difference of H_2 and D_2 as a function of pressure: $\Delta V/V = [V(\text{H}_2) - V(\text{D}_2)]/V(\text{D}_2)$; the data are from Refs [17, 18].

Lithium. A small but measurable difference in the lattice constants of ^6Li and ^7Li has been found in Ref. [14]. In Ref. [12], the equations of state of ^6Li and ^7Li were studied using an ultrasonic technique. Some of the results are shown in Fig. 5. It can be seen that the quantity

$$\frac{\Delta V}{V} = \frac{V(^6\text{Li}) - V(^7\text{Li})}{V(^7\text{Li})}$$

increases with pressure.

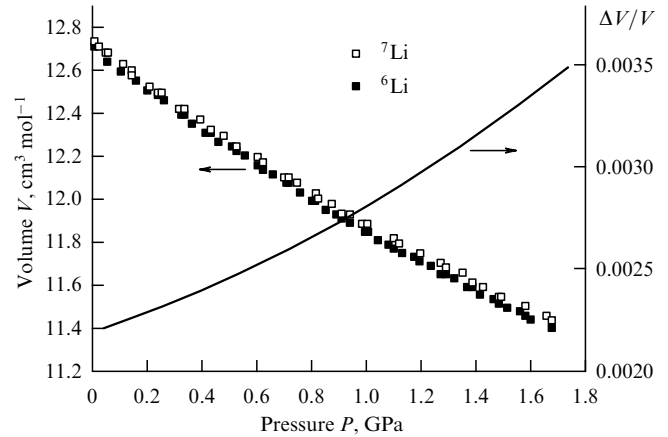


Figure 5. Compression isotherms and relative volume difference of ^6Li and ^7Li as functions of pressure at 77 K: $\Delta V/V = [V(^6\text{Li}) - V(^7\text{Li})]/V(^7\text{Li})$; the data are from Ref. [12].

Carbon (diamond). Recently, the lattice parameters of isotopically pure diamonds ^{12}C and ^{13}C were measured at various pressures [20]. We made use of the analytical representation of the results of Ref. [20] to calculate the quantities of interest here (Fig. 6). Again as in the case of lithium we observe a marked increase in the quantity

$$\frac{\Delta P}{K} \cong \frac{\Delta V}{V} = \frac{V(^{12}\text{C}) - V(^{13}\text{C})}{V(^{13}\text{C})}$$

with pressure. The previous conclusion about the increased role of quantum effects in diamond at high pressures was based on the study of Raman scattering from diamonds of ^{12}C and ^{13}C [21].

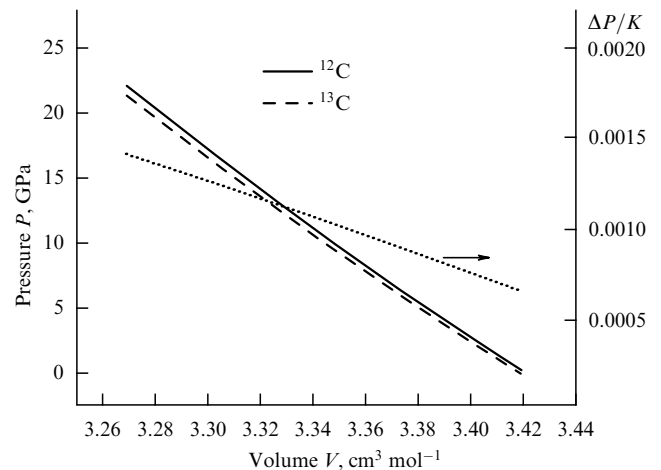


Figure 6. Compression of isotopically different diamonds at room temperature [20]. The quantity $\Delta P/K \cong \Delta V/V = [V(^{12}\text{C}) - V(^{13}\text{C})]/V(^{13}\text{C})$ is calculated from the data of Ref. [20].

Hydride and deuteride of lithium. Figure 7 presents compression isotherms for LiH and LiD and the quantity

$$\frac{\Delta P}{K} \cong \frac{\Delta V}{V} = \frac{V(\text{LiH}) - V(\text{LiD})}{V(\text{LiD})}$$

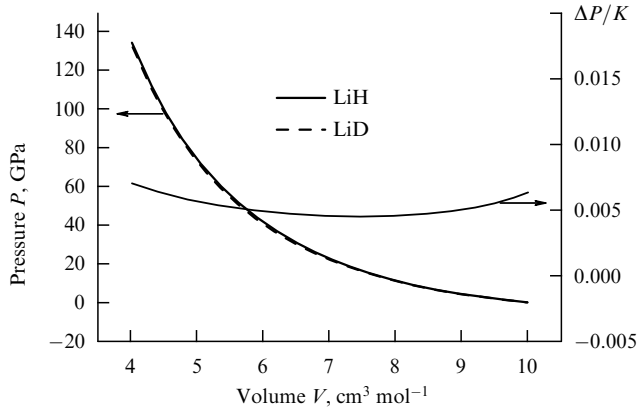


Figure 7. Compression isotherms for LiH and LiD at room temperature, based on data from Ref. [22]. The quantity $\Delta P/K \cong \Delta V/V = [V(\text{LiH}) - V(\text{LiD})]/V(\text{LiD})$ is calculated from the data of Ref. [22].

as a function of volume and resulting from calculations we performed based on the data of Ref. [22]. Note the high degree of compression achieved in this case. The dependence of $\Delta P/K$ on V has a nontrivial form, which fact, however — considering the measurement errors and those introduced by approximation procedures — most likely indicates the relative constancy of the quantity $\Delta P/K \cong \Delta V/V$.

4.2 Melting

Let us consider again the influence of quantum effects on the melting curve. We write the melting curve of a substance in the form $T_m \propto \rho^{n/3}$ or $T_m \propto V^{-n/3}$, where T_m is the melting temperature, ρ the density, and n the exponent describing the particle power-law interaction [8]. Clearly, the role of quantum effects will change along the melting curve in accordance with the relation

$$\frac{\lambda_T}{r} \propto T^{(2-n)/2n}, \quad (16)$$

where $\lambda_T = \hbar/(mkT)^{1/2}$ is the thermal de Broglie wavelength, and r is the interparticle distance.

From Eqn (13) it follows that as helium, hydrogen or a similar system with short-range repulsion melts, quantum effects disappear in the high-temperature limit, which is not inconsistent with the experimental data of Refs. [23, 24] (Figs 8 and 9).⁷ On the other hand, one cannot fail to see that the difference in melting temperature ΔT between helium isotopes changes unexpectedly little with pressure (Fig. 8).

At the same time, for a Coulomb system quantum effects occurring on melting must heighten with increasing pressure. This can be illustrated with numerical experiments on the melting of one-component plasma [6] (Fig. 10).⁸ As seen in Fig. 10, quantum effects lead to a maximum in the melting curve and ultimately exclude the crystalline phase for all temperatures. It is interesting to note that, according to Ref. [6], at the point where the melting curve of a crystalline plasma reaches a maximum (see Fig. 10) the thermal de Broglie wavelength is practically equal to the average interparticle distance, i.e. the relation $\lambda_T/r \cong 1$ holds.

⁷ There is evidence in the scientific literature that the isotope effect reverses sign during the melting of helium above 200 K [25].

⁸ One has to admit that in view of the ‘pathological’ properties of the one-component plasma model [26], any conclusions drawn from it must be looked upon with a bit of skepticism.

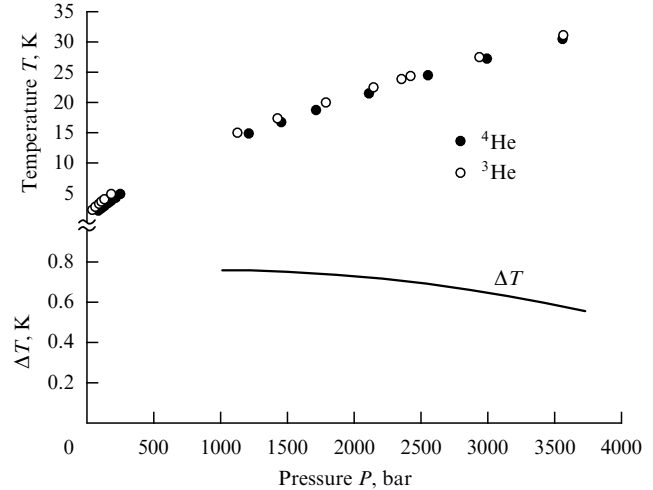


Figure 8. Melting curves of ^3He and ^4He [23]. ΔT is the difference between the melting temperatures of the isotopes at constant pressure.

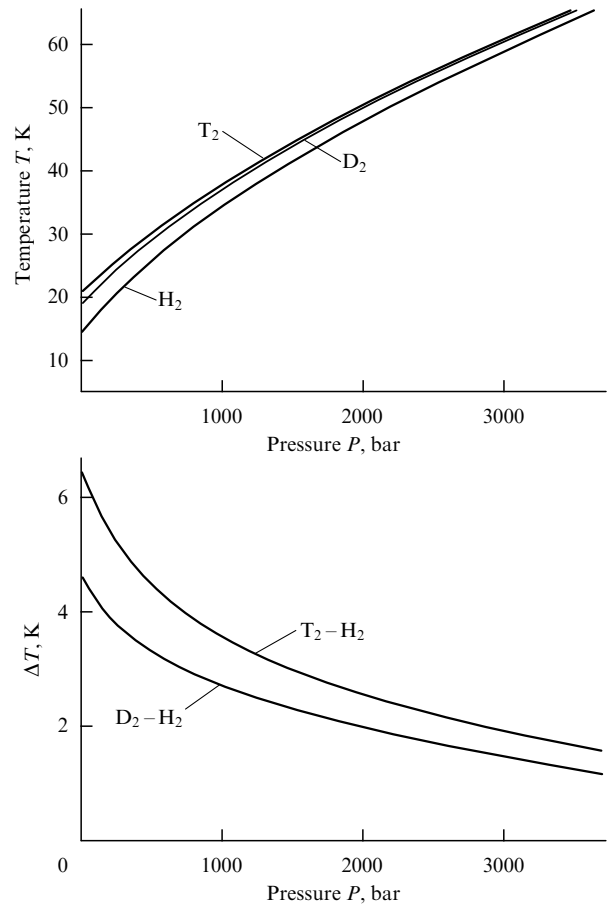


Figure 9. Melting curves of H_2 and D_2 [24]. ΔT is the difference between the melting temperatures of the isotopes at constant pressure.

5. Conclusions

The entire, if very limited, body of experimental and other material presented above generally agrees with the conclusions of Refs [2–4, 9] about the influence of pressure on quantum effects in systems with various types of interaction. We see, indeed, that quantum effects become stronger when

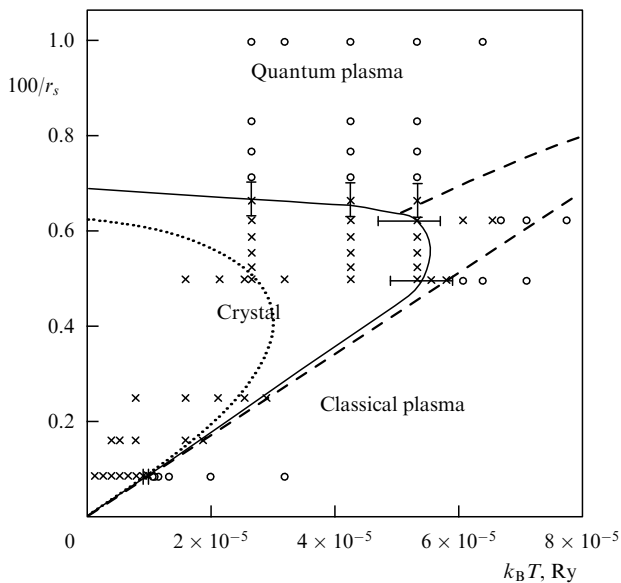


Figure 10. Phase diagram of a one-component quantum plasma [6].

metals and covalent crystals, i.e. systems with an essentially Coulomb interaction, are compressed.

On the other hand, quantum effects clearly decrease when van der Waals materials are compressed. The situation with ‘ionic’ LiH is unclear. One would expect, from the above, that a Coulomb substance will even melt at absolute zero provided high enough pressures are achieved. However, for those who reject the approaches dating back to Lindemann and do not content themselves with generalities, a clear picture of why this may happen is needed.

At first sight, the situation is quite clear. A naive view is that particles in a liquid move freely over the entire volume and hence — in accord with principle (1) — their kinetic energy is always less than that of particles in a crystal. But then the ‘zero-point’ energy of a liquid is always less than the ‘zero-point’ energy of a corresponding solid, and therefore an increase in the static energy on melting should be compensated for a corresponding decrease in the ‘zero-point’ energy. This is in fact not — or not precisely — so.

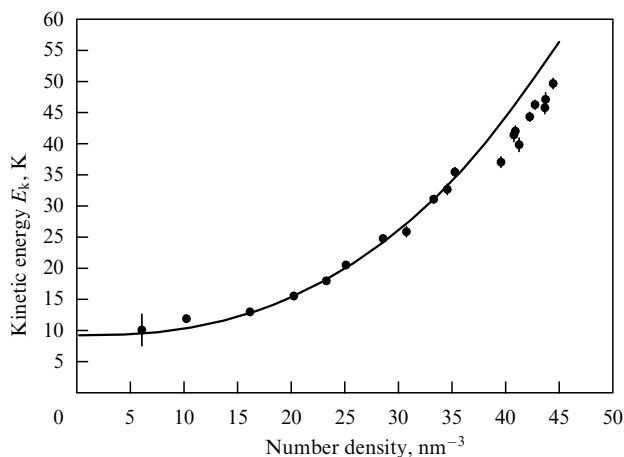


Figure 11. Average kinetic energy of ^4He atoms as a function of the density at 6.1 K [27]. The points at densities above 36.3 nm^{-3} correspond to solid helium.

The results of direct kinetic energy measurements on liquid and solid ^4He using deep inelastic neutron scattering are shown in Fig. 11 [27]. It is amazing but Fig. 11 demonstrates that the liquid helium kinetic energy is always higher than that of solid helium at one and the same density. This fact was first pointed out in Ref. [28].

Without discussing here what might cause this phenomenon (see Ref. [27] for more on that), we note in this connection that only a considerable amount of anharmonism in a liquid could reduce its zero-point energy compared with a crystal. One cannot rule out that the situation described in Refs [27, 28] is not universal and that the behavior of systems with a ‘soft interaction’ will correspond more closely to our intuition. One further point to be made is that, according to F London [29], in the ‘cold’ melting of helium the role of the zero-point energy is simply reduced to the expansion of the system to a larger volume, the liquid configuration of the particles becoming most favorable energetically (see Ref. [30] in this connection). Clearly, such a situation cannot be realized at pressures corresponding to the melting of a ‘Coulomb’ substance and, hence, the zero-point energy should play a more active role in that case.

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