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LONG RANGE FORCES BETWEEN ATOMIC IMPURITIES IN LIQUID HELIUM*

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Van der Waals or Casimir interaction between neutral quantum objects in their ground state is known to be universally attractive. This is not necessarily so when these objects are embedded in a polarizable medium. We show that atomic impurities in liquid helium may indeed realize repulsive forces, and even Van der Waals and Casimir forces with different signs.

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1. Introduction

Over the past twenty years, various experiments have dealt with atomic or molecular impurities embedded in liquid helium, superfluid or not. Solid helium have also been used. Successful theoretical models have been constructed to explain their properties. The main results have been reviewed recently in several articles [1–3]. Such systems realize almost ideally the situation of elementary quantum objects in a simple translation invariant and isotropic dielectric medium. Neglecting magnetic dipole interactions, long range interaction between those objects results from the electric charge density and electromagnetic field fluctuations, which produces the Van der Waals and Casimir forces [4,5]. A unified treatment of these forces has been given by Dzyaloshinskii *et al.* [6] in terms of dielectric constants and dynamic polarizabilities of the systems involved. Such forces are universally attractive for systems in their ground state in vacuum. It was pointed out in [6] that it is not necessarily so for unlike systems in a dielectric medium. This paper considers whether repulsive forces could arise between simple atomic

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or molecular impurities embedded in liquid helium. After a short discussion of the problem in Dyaloshinskii's approach, alkali atoms, rare gases, hydrogen atom and molecule are considered as candidates, their dynamic polarizabilities being available. We will show that repulsive interaction can indeed occur, and even cases in which Van der Waals and Casimir forces have different signs.

2. Basic formulae

The interaction of an atom or a molecule with other species is entirely determined by its dynamic polarizability. For long range forces, the dominant contribution comes from the electric dipole polarizability $\alpha(\omega)$ at frequency $\omega/2\pi$. Helium atom is weakly polarizable in the frequency range where other atoms have their eigenfrequencies. Liquid helium is also a low density material so that its dielectric constant $\varepsilon_{\mathrm{He}}$ is simply related to α_{He} and to the particle density ρ by

$$\varepsilon_{\mathrm{He}} = 1 + \rho\alpha_{\mathrm{He}}\,. \tag{1}$$

For zero temperature and pressure, $\rho = 0.0218 \times 10^{-24}\mathrm{cm}^{-3}$. Using its analytical properties, the function $\alpha(\omega)$ can be conveniently defined on the imaginary axis of ω. More precisely, we use as a variable ξ the imaginary part of ω in atomic units

$$\xi = \frac{\hbar\,\mathrm{Im}\,(\omega)}{2\mathrm{Ry}}\,, \tag{2}$$

where Ry is the Rydberg constant in energy units. A simple representation of helium polarizability $\alpha_{\mathrm{He}}(i\xi)$ is taken from reference [7].

$$\alpha_{\mathrm{He}}(i\xi) = 4\pi a_0^3 \frac{1.3853 + 1.21\xi^2 + 0.1028\xi^4}{1 + 1.992\xi^2 + 0.8362\xi^4 + 0.05272\xi^6}\,, \tag{3}$$

where a_0 is the Bohr radius.

Two impurities A_1 and A_2, characterized by their polarizabilities α_1 and α_2 are introduced in liquid helium. They replace respectively n_1 and n_2 helium atoms. The volume $V_1 = n_1/\rho$ occupied by the impurity A_1 is characterized by an average dielectric constant $\varepsilon_1 = 1 + \alpha_1/V_1$. According to Ref. [6], the force between two spheres with volumes V_1 and V_2 characterized by dielectric constants ε_1 and ε_2 separated by a distance R in a dielectric medium with a dielectric constant $\varepsilon_{\mathrm{He}}$ derives from a potential $U(R)$ which can be expressed as a function of *excess dielectric constants* $\varepsilon_1 - \varepsilon_{\mathrm{He}}$ and $\varepsilon_2 - \varepsilon_{\mathrm{He}}$. For distances shorter than the wavelength λ_0 associated with typical excitations of the media ε_1 and ε_2, $U(R)$ is an effective Van der Waals interaction

$$U(R) = -\frac{3(2\mathrm{Ry})}{\pi\ R^6} V_1 V_2 \int_0^\infty d\xi\ \frac{[\varepsilon_1(i\xi) - \varepsilon_{\mathrm{He}}(i\xi)][\varepsilon_2(i\xi) - \varepsilon_{\mathrm{He}}(i\xi)]}{(4\pi\varepsilon_{\mathrm{He}}(i\xi))^2} \tag{4}$$

$$= -\frac{3(2\mathrm{Ry})}{\pi\ R^6} \int_0^\infty d\xi\ \frac{[\alpha_1(i\xi) - n_1\alpha_{\mathrm{He}}(i\xi)][\alpha_2(i\xi) - n_2\alpha_{\mathrm{He}}(i\xi)]}{[4\pi(1 + \rho\alpha_{\mathrm{He}}(i\xi))]^2}\,. \tag{5}$$

For $R \gg \lambda_0$, $U(R)$ is an effective Casimir interaction

$$U(R) = -\frac{23\hbar c}{\pi\sqrt{\varepsilon_{\mathrm{He}}(0)}\ R^7} V_1 V_2 \frac{[\varepsilon_1(0) - \varepsilon_{\mathrm{He}}(0)][\varepsilon_2(0) - \varepsilon_{\mathrm{He}}(0)]}{(8\pi\varepsilon_{\mathrm{He}}(0))^2} \tag{6}$$

$$= -\frac{23\hbar c}{\pi\sqrt{\varepsilon_{\mathrm{He}}(0)}\ R^7} \frac{[\alpha_1(0) - n_1\alpha_{\mathrm{He}}(0)][\alpha_2(0) - n_2\alpha_{\mathrm{He}}(0)]}{[8\pi(1 + \rho\alpha_{\mathrm{He}}(0))]^2}\,. \tag{7}$$

Both formulae involve the *excess polarizabilities* $[\alpha_{\mathrm{A}} - n_{\mathrm{A}}\alpha_{\mathrm{He}}]$. The weak helium polarizability is α_{He} is multiplied by n_{A}, which in some cases is much larger than 1. It is clear that when A_1 and A_2 are identical, the quantity to integrate in the second hand of equation (4) is positive, as well as the bracket product in equation (6). Both interaction are attractive.

To achieve a repulsion for the Van der Waals force, two conditions must be fulfilled.

1. $[\alpha_j(i\xi) - \alpha_{\mathrm{He}}(i\xi)]$ should be negative for one of the impurities ($j =$ 1 or 2) over a significant range of ξ values. This means that the corresponding atom should be less polarizable in this frequency range than the displaced helium atoms.
2. the spectra of the two atoms A_1 or A_2 must also differ from another enough to make the product $[\alpha_1(i\xi) - \alpha_{\mathrm{He}}(i\xi)][\alpha_2(i\xi) - \alpha_{\mathrm{He}}(i\xi)]$ negative on a large frequency range.

For the Casimir force, the condition is simpler since only the static polarizabilities appears. It is repulsive when one of the impurity (and only one) is less polarizable than the replaced helium atoms, while the other is more polarizable.

We now consider various type of atoms to achieve such situations.

3. Alkali atoms

Alkali atoms are as much different as possible from helium atoms, and are thus good candidates. Alkali atoms are known to attract only weakly with liquid helium, the interaction potential being mainly repulsive. They may be pictured as making a kind of bubble in liquid helium. Its radius is approximately the core radius r_c of the He–A potential (A is a generic alkali atom). Hence they replace on the average $n_A = 4\pi r_c^3/3$ helium atoms. The dynamic polarizabilities of alkali atoms will be represented by the two-term Padé approximation given in [8]

$$\alpha_A(i\xi) = 4\pi\ F_1 \frac{1}{\xi_1^2 + \xi^2} + 4\pi\ F_2 \frac{1}{\xi_2^2 + \xi^2}\,. \tag{8}$$

Lithium, sodium and cesium atoms will be taken as examples. The relevant parameters are given in Table I.

TABLE I

Parameters used for alkali atoms in atomic units. The value of r_c is taken from Ref. [9], parameters for polarizability (8) is from Ref. [8].

Atom A	r_c	n_A	F_1	ξ_1	F_2	ξ_2
Li	10.4	15.2	0.0413	0.1564	0.748	0.0678
Na	10.75	16.8	0.986	0.0775	0.0	0.0152
Cs	13	29.6	0.459	0.1619	1.116	0.0534

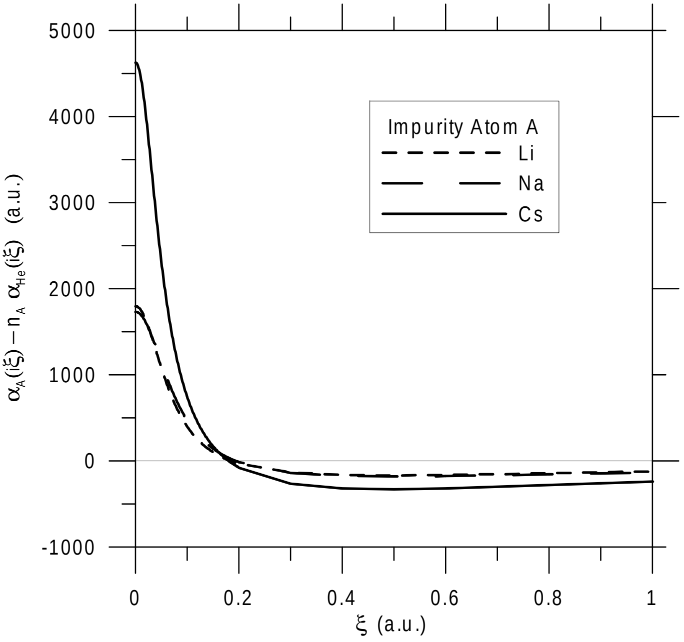

Fig. 1. Excess dynamic polarizabilities of Li, Na and Cs embedded in liquid helium (in atomic units, 1 a.u. $= a_0^3$) *versus* imaginary frequency defined by formula (2).

The excess dynamic polarizabilities of these alkali atoms in liquid helium are shown in figure 1. It is clear that the condition 1 is fulfilled from $\xi = 0.2$ up to ∞. However the spectrum of the two alkali are not much different and the product of the excess polarizabilities is mostly positive. The Van der Waals interaction between alkali impurities remains attractive. The same holds true for the Casimir interaction. Note that the excess dynamic polarizability integral from $\xi = 0$ to an upper limit is larger than 1.3 is negative for all alkali atoms. We will take advantage of this specific feature later on.

4. Rare gas atoms

We consider Ne and Xe as extreme examples, and Ar in between. Polarizabilities are taken from Kiaslyakov [7]. The structure of liquid helium around rare gas impurities has been studied by Dalfovo [11]. In contrast to the alkali atom case, the attraction potential produced by the rare gas atom is strong enough to produce a dense shell of helium atoms around its hard core. The radius of the hard core is also much smaller (about 5.4 a.u. for Ne, 7.4 a.u. for Xe). The overall balance is positive. There is more helium atoms around the impurity than before its introduction in the same region. Quantitatively, we get from reference [11] $n_{\mathrm{Ne}} = -2.4$ and $n_{\mathrm{Xe}} = -1.7$. The resulting excess dynamic polarizabilities are shown in figure 2. They are one order of magnitude lower than those of alkali atoms and always positive. They keep significant values up to large values of ξ, reflecting the high excitation energies of rare gases, in particular Ne. The effective interaction between rare gases in liquid helium are thus always attractive. Consider now the pair Cs-Ne. The product of their polarizabilities is nearly proportional to that of Cs and its integral can be negative. Table II gives the Van der

TABLE II

Computed Van der Waals coefficients C_6 for various alkali–rare gas pairs embedded in liquid helium, in atomic units (1 a.u. $= 2\,\mathrm{Ry}\, a_0^6$). The pairs with a negative coefficient achieve a repulsive Van der Waals interaction. For comparison, the right part of the table gives the similarly computed values of C_6 for the same species in vacuum.

	Ne	Ar	Xe	Ne	Ar	Xe
	in liquid helium			*in vacuum*		
Li	−6.5	16	89	42	171	404
Na	−7.7	15	93	47	188	440
Cs	2.3	70	242	90	362	857

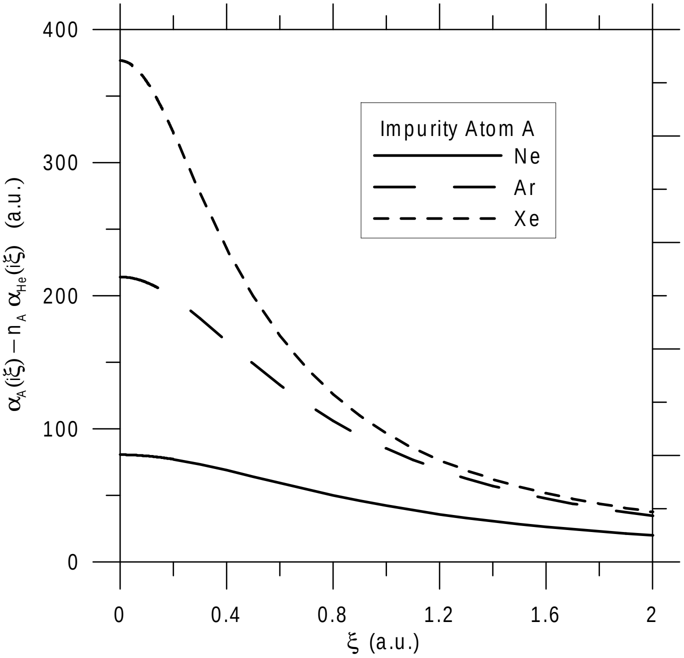

Fig. 2. Excess dynamic polarizability for Ne, Ar and Xe embedded in liquid helium.

Waals coefficients computed from formulae (4) using the excess polarizabilities shown in figures 1 and 2 for various alkali–rare gas pairs. Those pairs with a negative coefficient undergo a *repulsive* effective Van der Waals interaction. This is achieved for Li–Ne and for Na–Ne. The Van der Waals force nearly cancels also for Cs–Ne. This results from a non trivial cancellation of low and high frequency contributions. The static excess polarizabilities of all impurities considered up to now are positive. Thus the Casimir interaction is still *attractive*. The interaction force changes sign for distances intermediate between the excitation wavelengths of the alkali atom and those of the rare gas. This possibility was already mentioned in reference [6] as a theoretical one. We give here an actual example of such a peculiar behaviour. It must be kept in mind however that a revision of the alkali polarizabilities by about 10 percent could re-establish the positive sign of the effective Van der Waals constant.

5. Lighter elements

Light elements give simpler and more clear cut cases. We consider H atom, hydrogen molecule H_2 and the light isotope of helium ^{3}He. Their interaction with helium atoms are comparable [12]. According to this reference, the potential well may be modeled by Lennard–Jones potentials $V(R) = 4\epsilon\ [(\sigma/R)^{12} - (\sigma/R)^6]$ with parameters given in Table III. When introduced as an impurity in liquid helium-4, they occupy a larger volume than a ^{4}He atom because of their larger zero point motion. The number $n_{\rm A}$ of replaced ^{4}He atoms is also given in Table III. For ^{3}He, this number is

TABLE III

Lennard–Jones well (ϵ) and hard core radius (σ) for the interaction of light species with helium in atomic units, taken from reference [12]. The number $n_{\rm A}$ of replaced helium atoms is computed from the volume coefficient α drawn from references [14,15] by $n_{\rm A} = 1 + \alpha$.

	ϵ	σ	$n_{\rm A}$	
H	5.31E-5	6.047	5 [14]	3 [15]
H_2	8.75E-5	5.725	1.2 [14]	2.9 [15]
^{3}He	7.08E-5	5.007	1.3	

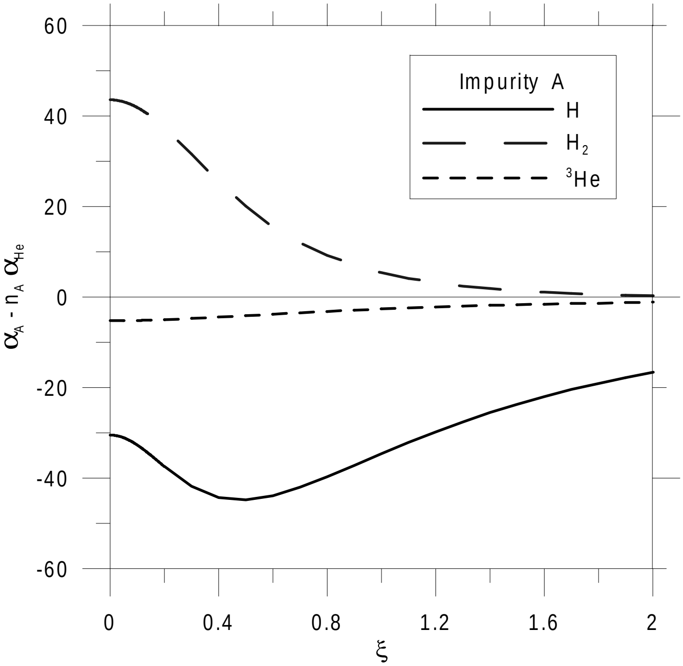

Fig. 3. Excess dynamic polarizabilities for H, H_2 and ^{3}He embedded in liquid helium.

deduced from the molar volume of dilute ^{3}He/^{4}He solutions [13]. For H and H_2, Kürten and Ristig [14] have computed values for the relative excess coefficient $\alpha = n_{\rm A} - 1$. Crude estimates by Marin *et al.* [15] are similar for H, but much larger for H_2. For the dynamic polarizability of H, formula (8) is used with $F_1 = 0.7350$, $\xi_1 = 0.3793$, $F_2 = 0.4268$, $\xi_1 = 0.6025$. For H_2, we use the polarizability computed by Caffarel *et al.* [16], rotationally averaged, and fitted by the following expression in atomic units $\alpha_{\rm H_2}(i\xi) = 4\pi\alpha_0\, \eta^2/(\eta^2 + \xi^2)$, with $\eta = 0.572$ and $\alpha_0 = 5.13$.

The resulting excess polarizabilities are shown in figure 3, using $n_{\rm A}$ from reference [14]. For H and ^{3}He, they are negative for all values of ξ. As a consequence, these atoms in liquid helium are repelled by any other impurity with a positive excess polarizability. This is expected for most closed shell atoms and molecules. Hydrogen molecules behave like rare gases for this choice of $n_{\rm A}$. With the larger value of reference [15], the excess polarizability is negative at high frequencies and positive up to $\xi = 0.4$. It is similar to alkali atoms. Hence the present uncertainty about $n_{\rm A}$ prevents drawing any definite conclusion for H_2.

Coming back to H and ^{3}He, various alkali atom impurities may interact differently with them because of their sign changing excess dynamic polarizabilities. One finds indeed that ^{3}He is weakly repelled by Cs, but attracted by Li and Na in the Van der Waals range. The Casimir force is repulsive in all cases. The same holds true for H with all alkali atoms: they *repel* in the Casimir distance range, but *attract* in the van der Waals range.

TABLE IV

Computed Van der Waals coefficients C_6 between H, ^{3}He and alkali atoms embedded in liquid helium. For comparison, the right part of the table gives the similarly computed values of C_6 for the same species in vacuum

	Li	Na	Cs	Li	Na	Cs
	in liquid helium			*in vacuum*		
H	24	26	41	65	71	139
^{3}He	0.24	0.3	−0.5	22	24	46

6. Conclusion

We have illustrated the fact that the effective force between two impurities in liquid helium results from a delicate balance of various contributions. They may be repulsive and eventually change their character at distances of several hundred nanometers. While the repulsive force between light elements and strongly attractive atoms or ions was already known in the helium physics community, we have pointed out that alkali atoms exhibit specific properties, with sign changing excess polarizabilities. Their interaction properties with other species are in some cases quite counterintuitive.

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