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REPULSIVE CASIMIR AND VAN DER WAALS FORCES: FROM MEASUREMENTS TO FUTURE TECHNOLOGIES

J. N. MUNDAY

Thomas J. Watson Laboratories of Applied Physics,
California Institute of Technology,
Pasadena, CA 91125, USA

FEDERICO CAPASSO

School of Engineering and Applied Sciences, Harvard University,
Cambridge, MA 02138, USA

By engineering the boundary conditions of electromagnetic fields between material interfaces, one can dramatically change the Casimir-Lifshitz force between surfaces as a result of the modified zero-point energy density of the system. Repulsive interactions between macroscopic bodies occur when their dielectric responses obey a particular inequality, as pointed out by Dzyaloshinskii, Lifshitz, and Pitaevskii. We discuss experimental verification of this behavior as well as a description of how this can be used to develop a scheme for quantum levitation. Based on these concepts, we discuss the possible development of a new class of devices based on ultra-low static friction and the ability to sort objects based on their dielectric functions.

Keywords: Casimir; van der Waals; Lifshitz; Levitation.

1. Introduction

The confinement of electromagnetic fields between material surfaces can result in a force between the latter due to quantum fluctuations of the former, which has many interesting features. First, this force represents a macroscopic manifestation of the quantum nature of the vacuum and is measurable using current experimental techniques. Second, a closer examination of several phenomena in nature shows strong evidence that adhesion, friction, wetting and stiction are fundamentally a result of these quantum fluctuations. Third, with the continued miniaturization of devices to the nanoscale, the ability to engineer the vacuum fluctuations between bodies may pave the way for improved device architectures, assembly methods, or functionalities. In this contribution, we will briefly discuss recent measurements of both long- and short-range repulsive forces, measurement schemes for future experiments, and technological opportunities that take advantage of the ability to modify these forces resulting from the confinement of vacuum fluctuations.

2. The Casimir-Lifshitz Force

The general expression for the force between two semi-infinite plates separated by a third medium as a result of the quantum fluctuations of the electromagnetic fields was first derived by Dzyaloshinskii, Lifshitz, and Pitaevskii [1]. In this formulation, the force between two uncharged surfaces, composed of either metals or dielectrics, is derived using the fluctuation-dissipation theorem. Because measurements of the Casimir force are usually compared to this generalized theory rather than the special case of ideal metals developed by Casimir, we refer to the resulting force as the Casimir-Lifshitz force.

Lifshitz's theory has various limiting forms depending on the materials involved and their separations. At very small separations (typically less than a few nm), Lifshitz's theory provides a complete description of the non-retarded van der Waals force. At larger separations, retardation effects give rise to a long-range interaction that in the case of two ideal metals in vacuum reduces to Casimir's result. Thus, both the Casimir force and the van der Waals force are of quantum electrodynamical (QED) origin, but the key physical difference is that in the Casimir case, the retarded nature of the interaction due to the finite speed of light cannot be neglected, as in the van der Waals limit. Retardation effects are actually dominant and lead to a change in the power law of the force with distance [2]. This is true for all materials (metals or dielectrics) when the propagation time of light between the bodies is greater than the inverse characteristic frequency of the materials [2], which for metals is the plasma frequency. The complete theory for macroscopic bodies is valid for any distance between the surfaces and includes, in a consistent way, both limits [1].

3. Origin of Repulsive Forces

As was demonstrated by Dzyaloshinskii, Lifshitz, and Pitaevskii in their seminal paper, the sign of the force depends on the dielectric properties of materials involved [1]. Two plates made out of the same material will always attract, regardless of the choice of the intermediate material (typically a fluid or vacuum); however, between slabs of different materials (here labeled 1 and 2) the force becomes repulsive by suitably choosing the intermediate liquid (labeled 3). Thus, by proper choice of materials, the Casimir-Lifshitz force between slabs 1 and 2 can be either attractive or repulsive. Specifically, the condition for repulsion is:

$$\epsilon_1(i\xi) > \epsilon_3(i\xi) > \epsilon_2(i\xi). \tag{1}$$

Here the dielectric functions ϵ_1, ϵ_2, and ϵ_3 of the materials (Fig. 1) are evaluated at imaginary frequencies [3]. Because they vary with frequency, it is conceivable that inequality [Eq. 1] may be satisfied for some frequencies and not for others. For various separations between the slabs, different frequencies will contribute with different strengths, which can lead to a change in the sign of the force as a function of separation (see for example Ref. [4]).

Fig. 1. Geometry of the setup. Two plates separated by an intermediate fluid.

In order to qualitatively understand the origin of these repulsive forces, we consider the following toy model (see Fig. 2) for the microscopic interaction of the bodies [5]. To first order, the force between the latter is dominated by the pair-wise summation of the van der Waals forces between all the constituent molecules. This additivity is a good approximation for rarefied media; however, the force between two molecules is affected in general by the presence of a third. Hamaker first used this approach in extending the calculations of London to the short-range interaction (i.e. the non-retarded van der Waals force) between bodies and in particular to those immersed in a fluid [6]. Using the previous subscript notation for the three materials and their constituent molecules and suitably choosing them so that their polarizabilities satisfy the inequality $\alpha_1 > \alpha_3 > \alpha_2$, we find the forces between the individual molecules, which are proportional to the product of the polarizabilites integrated over all imaginary frequencies, will obey: $F_{13} > F_{12} > F_{23}$ (Fig. 2). Thus, it is energetically more favorable for molecule 3 to be near molecule 1 than it is for molecule 2 to be near molecule 1. As more molecules of the same species are added to the system, molecules of type 3 will be strongly attracted to those of type 1, resulting in an increased separation for molecules of type 2 from those of type 1. In this way, Hamaker showed, that repulsive forces between two different materials immersed in a liquid are possible by calculating the total interaction energy between the bodies and the fluid as the separation between the bodies is varied. His calculations however were non-rigorous since they neglected non-additivity and retardation effects. When these are included, long-range repulsion between two bodies (materials 1 and 2) separated by a third (material 3) is predicted when their relative dielectric functions obey [Eq. 1]. Note that when the fluid has the largest dielectric function, the cohesive van der Waals interaction within the fluid will results in an attraction between its molecules that is larger than that between the molecules of the fluid and the plate, which leads to an attractive force between the two plates.

Several examples of material systems that obey [Eq. 1] exist in nature. One of the earliest triumphs of Lifshitz's theory was the quantitative explanation of the thickening of a superfluid helium film on the walls of a container [1, 7]. For that system, the dielectric function of liquid is intermediate between that of the

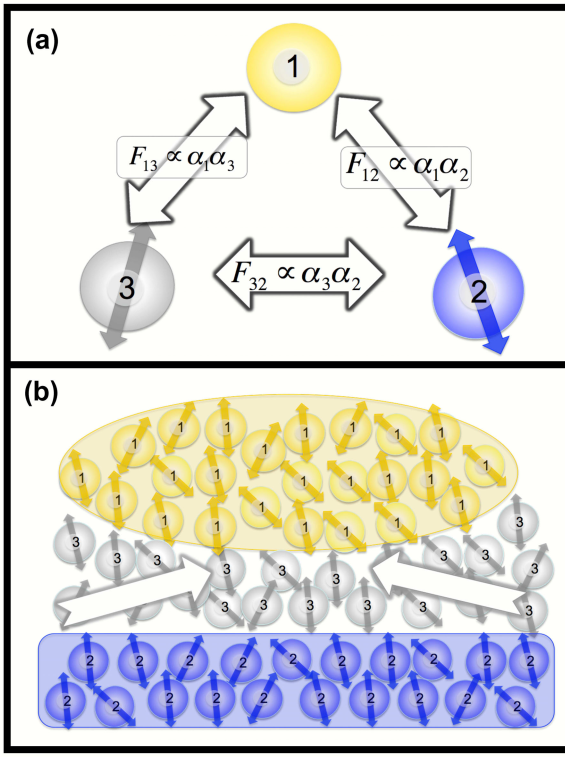

Fig. 2. Toy model of repulsive Casimir-Lifshitz forces. Repulsive forces can exist between two materials, schematically represented as an ensemble of molecules separated by a third, typically a liquid, with specific optical properties. (a) Three individual molecules will all experience attractive interactions. (b) For a collection of molecules, with $\alpha_1 > \alpha_3 > \alpha_2$, it is energetically more favorable for the molecules with the largest polarizabilities (α_1 and α_3 for this example) to be close, resulting in an increased separation between molecules of type 1 and type 2. For a condensed system, the net interaction between material 1 and material 2 is repulsive if the corresponding dielectric functions satisfy $\epsilon_1 > \epsilon_3 > \epsilon_2$, as consequence of the similar inequality between polarizabilities. Note that all the α's and the ϵ's need to be evaluated at imaginary frequencies (see text).

container and the surrounding vapor. Thus, it is energetically more favorable for the liquid to be between the vapor and the container, and the liquid climbs the wall. Of course the fact that the superfluid has also zero viscosity means that the fluid can easily spread and even flow out of a container depending on its height. Many other examples of this QED repulsion exist in the realm of wetting or non-wetting of a surface by a fluid, which to a first approximation, is governed by the same inequality for the dielectric functions.

4. Measurements of Repulsive Forces

Few material systems, consisting only of solids separated by a liquid, obey the inequality [Eq. 1] over a large frequency range; however, over the past decade there have been a limited number of force measurements for such systems. In this section we will briefly describe the commonalities and differences between these measurements. Although many methods have been developed to study surface forces [8, 9, 26, 27], the atomic force microscope (AFM) is one of the most widely used methods due to the versatility of material surfaces and surrounding environments that can be studied. Shortly after the development of the AFM [9], Ducker *et al.* performed one of the first measurements of the force between a colloidal sphere attached to a cantilever and a surface using AFM [10]. To our knowledge, all measurements of repulsive quantum forces to date (in both the retarded and non-retarded regimes) have used a similar setup; however, a variety of calibration methods and experimental techniques were used to probe different distance ranges with different materials.

Prior to our work, pervious experiments have shown evidence for short-range repulsive forces in the van der Waals regime [11–16]; however, there are many experimental issues that must be considered that, as our analysis below shows, were not adequately addressed in many of these experiments. For separations of a few nm or less, liquid orientation, solvation, and hydration forces become important and should be considered, which are not an issue at larger separations. Surface charging effects are important for all distance ranges. In order to satisfy [Eq. 1], one of the solid materials must have a dielectric function that is lower than the dielectric function of the intermediate fluid. One common choice for this solid material is PTFE (polytetrafluoroethylene), which was used in most experiments [12, 14–16]; however, as was pointed out in Ref. [12], residual carboxyl groups and other impurities can easily be transferred from the PTFE to the other surface, which complicates the detection and isolation of the van der Waals force. In a few experiments, the sign of the force did not agree with the theoretical calculation, which may be attributed to additional electrostatic force contributions [11, 12]. To avoid this problem, Meurk *et al.* performed experiments with inorganic samples [13]; however, the experimental configuration consisted of a sharp tip and a plate, which limited the surface separations to below 2 nm. For the determination of the cantilever force constant, either the Sader method [17] or the Cleveland method [18] was used in these experiments. The Sader method gives the spring constant of a cantilever based on the geometry of the cantilever and its resonance frequency, and the Cleveland method uses the resonance frequency shift of a cantilever upon the addition of masses to determine the spring constant. These methods lead to an additional 10-20% error in the determination of the force [19], which could be greatly reduced if a calibration method is performed that uses a known force for the calibration [20–22]. Finally, the determination of the absolute distance was often found by performing a fit of the experimental data to the presumed power law of the van der Waals force [12, 14–16]. Thus, the absolute surface separation could only be determined if one assumed that the measured force was only the van der Waals force and that it was described precisely by a $1/d^2$ force law.

In our recent experiment [23], we measured the long-range repulsive Casimir-Lifshitz force using (a) an improved force and distance calibration scheme, (b) methods to determine and reduce spurious electrostatic forces, and (c) spheres and cantilevers that would allow detection of weak forces at large surface separations. To calibrate the cantilever force constant and the surface separation at contact, a known force, the hydrodynamic force, was applied between the sphere and the plate [20–23]. This allows for in situ calibration and only assumes that the unknown force to be measured is independent of velocity. No assumption about the distance dependence of the force is made to determine the absolute surface separation. To ensure accuracy in the relative distance between the sphere and the plate, a linear variable differential transformer (Asylum Research MFP-3D) is used to control the piezo column, which advances the sphere toward the plate. This reduces hysteresis and nonlinearities inherent in piezoelectric transducers. To minimize electrostatic forces, we chose to use inorganic samples to avoid contamination by charge bearing groups often found with polymers. Further, we performed several investigations to ensure that electrostatic forces were negligible by removing stray charges and fields typically present near the apparatus [22] and performing electrostatic force microscopy on the samples [23–25]. Lifshitz's equation was computed using available dielectric data and corrections for surface roughness (as measured on both the sphere and the plate) to allow for an independent comparison of the theory and the experiment without any fitting parameters. This allowed us to conclude that theory and experiment are consistent within their numerical uncertainties and experimental errors, respectively.

5. Future Measurement Directions and Technological Opportunities

Although the AFM has been the instrument of choice for many surface force measurements, particularly for large surface separations, there is vast literature on other experimental techniques that may be of interest for measuring long-range surface forces in fluids. Techniques include the surface force apparatus (SFA) [8], total internal reflection microscopy (TIRM) [26], video tracking of colloids [27], and their various modifications. We will not discuss these schemes in detail but rather mention that these setups are capable of measuring forces either between macroscopic bodies [8] or between freely moving or confined particles above a surface [26, 27]. The latter offers the opportunity of observing quantum levitation by the Casimir-Lifshitz force without the support of a cantilever.

The ability to modify the Casimir-Lifshitz force opens the door to the possibility of engineering the potential energy landscape for particles based purely on their dielectric functions. Figure 3 shows an example of this behavior. With the appropriate choice of fluid, repulsive forces will occur for asymmetric configurations (Au-SiO2 in this case), while attractive forces will occur for symmetric configurations (Au-Au or SiO2-SiO2). By patterning a plate with these two different materials, one can study both non-additivity effects and the assembly and sorting of particles based solely on their dielectric functions. Similar sorting and aggregation effects have been observed

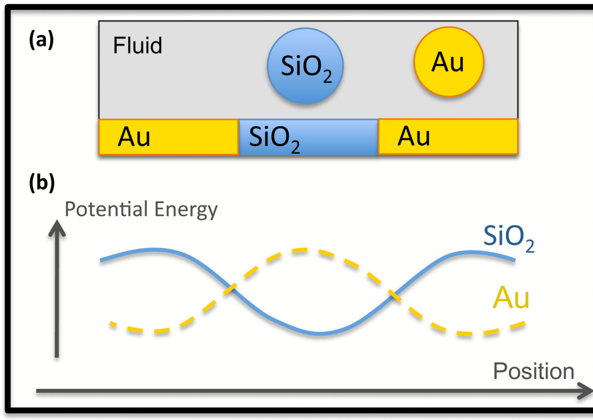

Fig. 3. Schematic of a method aimed at engineering the potential energy landscape of quantum fluctuations. (a) Two spheres made of different materials immersed in a fluid above a plate, which is also composed of two materials. The fluid and the two materials satisfy the inequality of dielectric functions discussed in the text [Eq. 1] (b) Schematic of the potential energy for both the Au (dashed) and the SiO2 (solid) spheres as a function of position for a fixed height above the plate. Each sphere experiences a different minimum energy configuration.

in the thermodynamic Casimir effect, which is related to classical density fluctuations [28]. The ability to achieve both attractive and repulsive Casimir-Lifshitz forces could be of significant interest technologically as well. One technique might be to develop ultra-sensitive force and torque sensors by counterbalancing gravity to levitate an object immersed in fluid above a surface without disturbing electric or magnetic interactions. Based on this idea, we proposed several devices that would be free to rotate or translate with virtually no static friction [29, 30]. Recent friction measurements have shown that ultralow friction can be obtained in such a configuration [16]. While dynamical damping due to viscosity will put limits on how quickly such a device can respond, in principle even the smallest translations or rotations can be detected on longer time scales. Thus, force and torque sensors could be developed that surpass those currently used. Other recent proposals include the ability to tune chemical reactions [31] and the self-assembly of colloidal scale devices [32, 33] based, at least partially, on manipulating the Casimir-Lifshitz forces.

6. Conclusion

Conclusive experiments have been discussed that demonstrate that the sign of the Casimir-Lifshitz force can be changed by an appropriate choice of materials. Recent proposals and measurements suggest that the role of the Casimir effect may be important for future technologies based on engineering the boundary conditions imposed on the ever-fluctuating electromagnetic fields.

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