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To cite this article: G. Torricelli et al 2011 EPL 93 51001

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Casimir force between a metal and a semimetal

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received 12 October 2010; accepted in final form 11 February 2011 published online 11 March 2011

PACS 12.20.Fv – Quantum electrodynamics: Experimental tests
 PACS 78.20.Ci – Optical properties of bulk materials and thin films: Optical constants (including refractive index, complex dielectric constant, absorption, reflection and transmission coefficients, emissivity)
 PACS 07.79.Lh – Scanning probe microscopes and components: Atomic force microscopes

Abstract – We present here measurements of the Casimir force gradient in the 60–300 nm range using a commercial Atomic Force Microscope operating in Ultra High Vacuum (UHV). The measurements were carried out in the sphere-plate geometry between a Au sphere and plates consisting of two different classes of material, that is a metal (Au) and a semimetal (HOPG). The variation in the optical properties of the materials produces clearly observed differences in the Casimir force as predicted by calculations based on the quantum theory of optical networks and the Lifshitz theory.



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Introduction. - The Casimir force is a remarkable mechanical effect between two macroscopic bodies induced by quantum fluctuations of the electromagnetic field [1]. It can be described in an intuitive way as the difference in radiation pressure of virtual photons outside and inside a cavity formed by two mirrors, resulting in an attractive force between them [2]. The original prediction by HBG Casimir in 1948 considered the ideal case of two perfectly reflecting and flat parallel mirrors at zero temperature [1]. Within this assumption, which excludes any material-dependent properties, the force depends only on fundamental constants and the geometry of the cavity. Since the original work, there has been an intense theoretical effort to describe the force for real materials considering the actual dielectric properties of the two mirrors and the intervening media [3–14]. It was realized that in the limit of small separation the Casimir force tends to the more familiar van der Waals force. These more realistic descriptions provide specific predictions that can be compared with high-precision measurements and they also predict how to modify the magnitude of the force. In 1997, Lamoreaux opened a new era of experiments using a torsional balance^[15] device 40 years after the pioneering work by Sparnaay [16]. Since then many experiments have been carried out, demonstrating

the renewal in interest in the measurement of the Casimir force [17–34]. As for the Lamoreaux measurement, most of these experiments were performed using metallic-coated surfaces [17–22,24–29,31]. The use of metallic surfaces was motivated by the need to prevent charge accumulations that can occur in non-conductive materials. The electrostatic interaction thus generated can affect the precision of Casimir force measurements. Using gold-coated surfaces, Decca and co-workers could measure the Casimir force with a claimed experimental error of 0.19% at the shortest distance of measurement [26]. Also, ways of tailoring the Casimir force have been investigated. For example, it has been demonstrated that by using sufficiently thin metallic films, the Casimir force can be significantly reduced compared to bulk metallic materials [25]. Prior to this measurement, there was a first attempt to tailor the Casimir force using hydrogen switchable mirrors [23]. Also, de Man and co-workers have demonstrated that the interaction between a gold sphere facing a gold plate is twice as large as the same sphere facing a conductive oxide such as Indium Tin Oxide (ITO) [32]. Measurements on semiconducting materials such as silicon [35], silicon membrane [36] and germanium [33] have also been investigated. Finally, the possibility of switching the force with a change of 20% has now been demonstrated using phase change materials [34]. The van der Waals and Casimir forces play a key role in Micro and

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Fig. 1: (Colour on-line) Dielectric constant at imaginary frequencies for Au sample (dotted orange line), and for the extraordinary (dashed grey line) and ordinary (black line) rays for HOPG.

Nano-Electromechanical Systems (MEMS/NEMS) [20,37]. Indeed, dispersion forces can induce non-linear behaviour in such systems [38]. Irreversible phenomena such as stiction or pull-in due to mechanical instabilities are observed when scaling down devices [39,40]. Therefore the ability to modify the Casimir interaction can impact strongly on the development of MEMS/NEMS. Here we report the Casimir force gradient measured between a Au sphere and a semimetallic plate of Highly Oriented Pyrolytic Graphite (HOPG). The measurement is compared with the force gradient between two Au surfaces. These two materials are known to have good conductivity at room temperature preventing charge accumulation, which could induce an electrostatic force in addition to the dispersion force. The measurements were performed in UHV and although this is not necessary for the relatively inert materials used in this study it does completely eliminate problems associated with, for example, capillary forces.

Theoretical estimate. – A significant contrast in the force can be expected only for materials with dielectric responses that differ over a wide range of frequencies [25,32,33,35,41]. Figure 1 shows the dielectric functions at imaginary frequencies, determined using the Kramers-Kronig relation. The optical data for Au was measured in the range [0.05–10] eV by J. A. Woollam Co., Inc. [42] on the actual sample used in this experiment, and was extrapolated to low frequencies, using a Drude model with a plasma and relaxation frequency of $\omega_p = 8.47 \text{ eV}$ and $\omega_r = 0.073 \text{ eV}$, respectively, which were obtained from a fit to the data. The calculation was performed using the method based on the Lifshitz theory [3,6,43]. HOPG is a uniaxial material therefore its tensor of permittivity is given by

$$\varepsilon_{ij} = \begin{bmatrix} \varepsilon_{xx} & 0 & 0\\ 0 & \varepsilon_{yy} & 0\\ 0 & 0 & \varepsilon_{zz} \end{bmatrix},$$
(1)

where $\varepsilon_{xx} = \varepsilon_{yy} \equiv \varepsilon_{\perp}$, $\varepsilon_{zz} \equiv \varepsilon_{\parallel}$. ε_{\perp} measures ordinary (o), while ε_{\parallel} measures extraordinary (e) ray properties. Optical data for graphite is available in the range $[10^{-5}-10^5]$ eV [44–46]. In the presence of the HOPG, the Casimir force formula has to be modified. In the case of parallel plates, one of them being a uniaxial crystal with a symmetry axis normal to its surface, the Transverse Electric (TE) and Transverse Magnetic (TM) polarizations can be considered separately. In the Casimir calculations the anisotropy shows up only in the reflection coefficients of graphite plate given by [47]

$$r_{gr}^{TM}(i\xi) = \frac{\varepsilon_{\perp}k_z - \sqrt{\frac{\varepsilon_{\perp}}{\varepsilon_{\parallel}}k_{\parallel}^2 + \varepsilon_{\perp}\frac{\xi^2}{c^2}}}{\varepsilon_{\perp}k_z + \sqrt{\frac{\varepsilon_{\perp}}{\varepsilon_{\parallel}}k_{\parallel}^2 + \varepsilon_{\perp}\frac{\xi^2}{c^2}}},$$
(2)

$$r_{gr}^{TE}(i\xi) = \frac{k_z - \sqrt{k_{\parallel}^2 + \varepsilon_{\perp} \frac{\xi^2}{c^2}}}{k_z + \sqrt{k_{\parallel}^2 + \varepsilon_{\perp} \frac{\xi^2}{c^2}}},$$
(3)

where $k_z = \sqrt{k_{\parallel}^2 + \frac{\xi^2}{c^2}}$.

The force was calculated within the proximity force approximation. No corrections other than the finite conductivity correction were applied here.

Experiments and results. - The main characteristics of the experimental set-up are described in ref. [48] in which there is also a full description of our experimental methodology. The experiments were performed at room temperature in UHV $(5 \times 10^{-11} \text{ mbar})$ using a commercial VT STM-AFM manufactured by Omicron nanotechnology. The probe consisted of a $20\,\mu\text{m}$ diameter polystyrene sphere attached at the end of a tipless silicon cantilever (Mikromasch [49]). The sphere was coated with a 5 nm adhesion layer of Cr and a 100 nm thick Au layer. The gold sample was a 100 nm thick layer evaporated in UHV onto a silicon wafer after deposition of a 5 nm thick Cr adhesion layer. The HOPG sample was obtained from PI-KEM Ltd. [50] and has a mosaic spread angle of 0.8° . It was cleaved just before introduction to the UHV system. AFM measurements on an area of $4 \times 4 \,\mu m^2$ gave an RMS roughness of 1.7 nm for the Au sample while the HOPG exhibited atomically flat terraces with few monoatomic steps. Therefore, in the range of distances probed in this work, the influence of roughness is negligible compared to the change in force produced by the different dielectric properties [29]. The force gradient between the sphere and the plate was measured in frequency modulation mode [31]. In this mode the cantilever vibrates at its resonant frequency $f_0 = 55.5 \,\mathrm{kHz}$ and the interaction between the sphere and the plate induces changes in its resonant frequency. In the linear regime the force gradient $\nabla F(z)$ is proportional to the frequency shift Δf :

$$\Delta f = \frac{-f_0}{2k} \nabla F(z), \tag{4}$$

where z is the distance between the sphere and the surface. It can be expressed as the sum of the z-piezo displacement



Fig. 2: (Colour on-line) Variations of V_0 with the distance. Each curve corresponds to a different position on either the Au plate (orange dots) or the HOPG plate (black dots).

and the distance z_0 at which the acquisition starts. Therefore a quantitative analysis would require a precise determination of the spring constant k of the cantilever and z_0 . The force was calibrated using the electrostatic interaction which can be controlled by applying a bias voltage V on the sphere. The frequency shift generated by an electrostatic interaction in the limit that the radius R of the sphere much bigger than z is

$$\Delta f = \frac{f_0}{2k} \frac{R\pi\varepsilon_0 (V - V_0)^2}{z^2},\tag{5}$$

where ε_0 is the vaccuum permittivity and V_0 is the contact potential difference. Before starting data acquisition V_0 was measured at the distance $z = z_0$ which here, is 6 nm above the closest distance of measurement. However, as also observed in previous studies, V_0 varies with the distance [33,34,51–54,]. This variation can be extracted from measurements at three different voltages (here, $\Delta V = V - V_0 = \pm 0.5$ V and $\Delta V = 0$ V) [48]. To simplify the discussion, we set the origin of the potential as $V_0 = 0$ at $z = z_0$. ΔV refers to its value at $z = z_0$. These three measurements are also used to determine kand z_0 . Moreover, $\Delta V = 0$ V corresponds to the measurement of the Casimir force gradient. For each sample the measurement has been performed in different areas (7 for Au and 6 for HOPG). These different regions investigated were for both samples spread over several mm. Moreover for each position, the measurement was repeated 40 times and prior any other analysis consecutive runs are averaged. Between each acquisition a feedback loop is applied in order to keep z_0 constant. Variations of V_0 are shown in fig. 2. Each line corresponds to a measurement at a different position on either HOPG sample (black dots) or Au sample (orange dots). The error on V_0 is dominated by the experimental error during its first determination before the data acquisition. For each position, the error can be then directly determined from the deviation



Fig. 3: (Colour on-line) Measurements of the Casimir force gradient between a Au sphere and a Au plate (orange dots) or a HOPG plate (black dots). The black line (resp. red line) is the force gradient calculation of the HOPG plate (resp. Au). Upper inset: force gradient difference between the Au-Au and Au-HOPG measurement. The experimental difference (grey line) is compared with the theory (black line). Lower inset: power law n of the force gradient as determined from the Au-Au measurements (orange dots), the Au-HOPG measurements (black dots) and from the theory (orange and black lines).

between the experimental value of V_0 at the distance z_0 shown in fig. 2 with the theoretical value that is here $V_0 = 0$ V. The measurement of the Casimir interaction was performed by compensating the voltage only at z_0 . Therefore the electrostatic contribution induced by changes of V_0 has to be subtracted afterwards. However, variations of V_0 are of the same order of magnitude for all measurements. At short distances the largest value found for V_0 is $-5 \,\mathrm{mV}$ while at 300 nm it is mainly below 10 mV apart for one position on the HOPG for which V_0 is almost $25 \,\mathrm{mV}$. The estimated remaining electrostatic interaction is much smaller than the Casimir interaction. Indeed, even considering the two extreme values of V_0 , the remaining electrostatic contribution is 4 orders of magnitude smaller than the Casimir contribution at short distances while at 300 nm the electrostatic contribution is still two orders of magnitude smaller. Therefore, variations of V_0 have a negligible contribution and do not affect the comparison of the Casimir force gradient between both materials. The values of z_0 and k were determined by fitting the average of the two experimental curves for $\Delta V = \pm 0.5$ V at $z = z_0$ after subtraction of the curve $\Delta V = 0$ V at $z = z_0$. Thus the parameters are calibrated independently of the variation of V_0 [48]. Figure 3 shows the Casimir force gradient measurements for both samples Gold (orange) and HOPG (dark grey). The remaining electrostatic contribution has been subtracted even though it is negligible. All curves are superimposed in order to show the reproducibility of the measurements over the different positions. The feedback parameters to maintain z_0 were $\Delta f = 150 \,\text{Hz}$ with

 $\Delta V = -0.5 \text{ V}$ which corresponds to $z_0 = 59.6 \text{ nm} \pm 0.5 \text{ nm}$ for Au-Au measurements and $z_0 = 58.7 \text{ nm} \pm 0.8 \text{ nm}$ for Au-HOPG measurements. The deviation is given here by the measurements in different areas of the sample and not the standard deviation of the 40 consecutive measurements taken at the same position which is always below $0.05\,\mathrm{nm}$. Even though the feedback parameters are the same for both samples, z_0 is different. A smaller z_0 for the HOPG is consistent with a weaker Casimir interaction compared to Au. A clear separation of the measured force gradient between both materials is observed. The theoretical curves are also shown and are in good agreement with the experimental data with an accuracy of between 2-12%. Moreover, the agreement could be improved by shifting the experimental curves to smaller distances. A shift of 1.2 nm for Au and 0.8 nm for HOPG would give the best agreement which could be partially explained by the use of an approximate formula for the capacitance which here over estimates z_0 by 0.5 nm [32, 48]. The upper inset of fig. 3 shows that the experimental difference between the two samples (green line), which varies from 25% at short distances (~ 50 nm) up to almost 35% at larger distances (> 200 nm). This difference is compared with the theory (dark grey line), which is consistent with the measured data although the measured difference is 5%bigger than the theoretical one. A source of the discrepancy can be the dielectric data for HOPG. Actual values could be different to those taken from the literature for the calculation. As has already been pointed out, a very accurate measurement would require the measurement of the optical constant of the actual sample used [55]. Moreover, differences in the calculated dispersion force can be obtained even between different Au films [56,57]. Finally, for any signal S(z) which obeys to a law such as $S(z) = Az^n$ with A a constant, the power law n can be determined by the relation

$$n = \frac{\mathrm{d}\log(S(z))}{\mathrm{d}\log(z)}.$$
 (6)

We applied this formula to experimental data and to theoretical curves for both samples in order to obtain an effective power law for the Casimir force gradient. However, this method needs a numerical derivative which can diverge when the noise becomes significant compared to the signal. Thus, a smoothing of the curve prior to the derivative has been performed. The result is shown in the lower inset of fig. 3. For both samples a fast drop below 90 nm was observed. Theoretically, n should be smaller than 3, which corresponds to the power law of force gradient of the non-retarded van der Waals limit between a sphere and a plane. Moreover, n has been also determined for electrostatic curves (not shown here). In this case a fast drop was also observed at the same distance. In the case of the electrostatic interaction n should be constant and equal to 2. The average value of n considering only distances beyond 100 nm is 2.02 ± 0.05 . Thus one can notice, as in ref. [53],

that no anomalous scaling of the electrostatic interaction is observed within this range. The z-dependence of the electrostatic interaction has been a concern since anomalies in the scaling has been reported by Kim et al. [52]. Therefore we believe that these fast variations at short distance are related to an experimental artefact¹. However, these variations are observed for both samples and therefore do not affect the qualitative comparison between both samples. Theoretically n is bigger for the Au-HOPG interaction than for the Au-Au interaction below 190 nm. At 190 nm there is a cross over between the two curves, and beyond that distance the theoretical n of the Au-Au interaction becomes larger than for the HOPG interaction. Experimental values of n are in agreement with the theoretical values although despite the smoothing, the data are not sufficiently precise to observe the difference between both samples. To compare more precisely the power law between the experiment and the calculation, we determine the average power law \overline{n} as done in [58] by fitting with a function proportional to $z^{-\overline{n}}$. This method has the inconvenience of losing the information of variations of n with z but it is not affected by the divergence implied by derivative processes. For this comparison, the experimental and theoretical data were between 100-300 nm. Because *n* varies with the distance, another range of distance would result in another value. The result of the fit give a theoretical $\overline{n} = 3.62$ for HOPG and $\overline{n} = 3.60$ for Au while experimentally we found $\overline{n} =$ 3.60 ± 0.04 for HOPG and $\overline{n} = 3.62 \pm 0.03$ for Au. Thus there is a good agreement between the experiment and the calculation. This good agreement suggests that the zdependence of the force gradient for the distances probed here can be satisfactorily explained by considering only the finite conductivity of the materials. However, a more complete description would require the temperature to be taken into account. Within a zero-temperature assumption n varies from 3 at short separation which correspond to the force gradient in the sphere plane geometry in non-retarded van der Waals limit up to 4 in the purely retarded regime in which limit the interaction is sample independent [5]. In [58] the non-retarded van der Waals regime in the sphere plate geometry using gold materials was experimentally determined below 18 nm. While in [5] the van der Waals regime was theoretically determined below 4 nm. At room temperature n reaches a maximum value which is always smaller than 4 at about $1\,\mu\mathrm{m}$ separation and then drops to reach 3 for very large $distances^2$.

¹For instance, piezoelectric materials may exhibit creep effect that could for the first points measured induce an actual z displacement smaller than the expected displacement.

²Variation of the effective power law of the Casimir energy between two gold mirrors was presented by Professor Adrian Parsegian at the workshop "Casimir Forces and Their Measurement" at Yale University in August 2009. A. Parsegian performed his computation using all four dielectric terms from table L2.4.1 in [13].

Conclusion. – In conclusion we have measured the Casimir force gradient between a semimetal and a metal. We have demonstrated that at room temperature the Casimir force gradient between a gold sphere and a HOPG surface is 20–30% smaller than for two gold surfaces. Also, we have found a very good agreement between the power law of the experimental curves and the theoretical power law. HOPG is a commonly used material, which has a high conductivity at room temperature, a very low roughness at the atomic level and is inert. We believe that HOPG and more generally graphite films can therefore be a good alternative for gold layers for use in MEMS devices which require van der Waals and Casimir interactions to be as small as possible.

* * *

We gratefully acknowledge the European Contract No. STRP 12142 NANOCASE and the EPSRC grant EP/F035942/1 for financial support and the ESF Casimir network which facilitated useful discussions.

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