## Do the precise measurements of the Casimir force agree with the expectations?

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## Abstract

An upper limit on the Casimir force is found using the dielectric functions of perfect crystalline materials which depend only on well defined material constants. The force measured with the atomic force microscope is larger than this limit at small separations between bodies and the discrepancy is significant. The simplest modification of the experiment is proposed allowing to make its results more reliable and answer the question if the discrepancy has any relation with the existence of a new force.

The Casimir force [1] between closely spaced macroscopic bodies is the effect of quantum electrodynamics (QED) and for that reason could be predicted very accurately. In the rigorous Lifshitz theory [2, 3] the force is defined by the optical properties of used materials. Knowledge of these properties is the weakest element in the theory restricting the accuracy that can be achieved. Though the measurement of the Casimir force is not the best way to test QED, such experiments are of great importance because they are sensitive to the presence of new fundamental forces [4] predicted in many modern theories (see, for example, [5] and references therein). To distinguish a new force from the background, we should be able to calculate the Casimir force with a precision better than the experimental one. In the series of recent experiments this force has been measured with the torsion pendulum (TP) [6] in the range of distances  $0.6-6 \ \mu m$  and with the atomic force microscope (AFM) [7, 8] in the range  $0.1 - 0.9 \ \mu m$ . The corresponding precisions were 5% and 1%, respectively.

The force per unit area between parallel plates arising as a result of electromagnetic fluctuations at nonzero temperature T is generalized by the Lifshitz theory [3],

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where the plate material is taken into account by its dielectric function at imaginary frequencies  $\varepsilon(i\zeta)$ :

$$F^{pl}(a) = \frac{kT}{\pi c^3} \sum_{n=0}^{\infty} \zeta_n^3 \int_{1}^{\infty} dp p^2 \left\{ \left[ G_1^2 e^{2p\zeta_n a/c} - 1 \right]^{-1} + \left[ G_2^2 e^{2p\zeta_n a/c} - 1 \right]^{-1} \right\}.$$
 (1)

Here "prime" means that n = 0 term is taken with the coefficient 1/2, a is the distance between bodies and

$$G_1 = \frac{p+s}{p-s}, \quad G_2 = \frac{\varepsilon (i\zeta_n) p+s}{\varepsilon (i\zeta_n) p-s},$$
  
$$s = \sqrt{\varepsilon (i\zeta_n) - 1 + p^2}, \quad \zeta_n = 2\pi n k T/\hbar.$$
(2)

The Casimir result  $F_c^{pl}(a) = \pi^2 \hbar c/240 a^4$  [1] is reproduced from (1) in the limit  $\varepsilon \to \infty$  and  $T \to 0$ . The function  $\varepsilon (i\zeta_n)$  cannot be measured directly but can be expressed via imaginary part of the dielectric function on the real axis with the help of the dispersion relation

$$\varepsilon(i\zeta) - 1 = \frac{2}{\pi} \int_{0}^{\infty} d\omega \frac{\omega Im\varepsilon(\omega)}{\omega^2 + \zeta^2}.$$
(3)

Information on  $Im\varepsilon(\omega)$  can be extracted from the data on reflectivity and absorptivity of electromagnetic waves for a given material.

In the experiments the force is measured between metalized disc and sphere because for two plates it is difficult to keep them parallel. For this configuration (1) has to be modified with the help of the proximity force theorem (PFT) [9] which is true for  $R \gg a$ , where R is the radius of curvature of the spherical surface. Applying PFT to (1) one can find the force between sphere and plate as  $2\pi R \int F^{pl}(a) da$ . The integration gives

$$F(a) = -\frac{kTR}{c^2} \sum_{n=0}^{\infty} \zeta_n^2 \int_1^\infty dpp \ln\left[\left(G_1^{-2}e^{-2p\zeta_n a/c} - 1\right)\left(G_2^{-2}e^{-2p\zeta_n a/c} - 1\right)\right].$$
 (4)

This expression differs from those used in [8] and [12] in two respects. First, in the cited papers the integration connected with the PFT was not done analytically that complicated numerical analysis. Second, the zero temperature limit has been taken when one can change the sum over n in (4) by the integral over  $\zeta$ . This limit was also considered in [13], though the PFT integral was evaluated explicitly. It seems a reasonable approximation at small separations because the temperature correction is proportional to  $(kTa/\hbar c)^3$  [10] and is small. However, one should remember

that this correction has been found in the limit of ideal conductor  $\varepsilon \to \infty$ . For a real conductor it can behave as  $kTa/\hbar c$  and be important. We have computed the force according to (4) and with the integral instead of the sum and found that the difference at the smallest distances tested in the AFM experiments exceeds 4 pN in contrast with the conservative estimate for the experimental errors 2 pN [8].

In the AFM experiments an additional  $Au_{0.6}Pd_{0.4}$  layer of 20 nm [7] or 8 nm [8] thick was on the top of Al metallization of the bodies to prevent aluminum oxidation. It has to be included into consideration. This layer is transparent for the electromagnetic waves with high frequencies  $\sim c/a$  since the absorption is proportional to  $Im\varepsilon(\omega)$  which is small for  $\omega \sim c/a$  and for this reason the layer was ignored in [7, 8]. However, the force depends on  $\varepsilon(i\zeta)$  for which the low frequencies dominate in (3) because of large  $Im\varepsilon(\omega)$  and that is why we cannot neglect the Au/Pd layer. To take it into account, one has to generalize expression for the force (1) to the case of layered bodies. Suppose that the top layer has the thickness h and its dielectric function is  $\varepsilon_1$ . The bottom layer is thick enough to be considered as infinite and let its dielectric function be  $\varepsilon_2$ . The method described in [3] for deriving Eq.(1) can be easily generalized for layered plates. We have to add only the matching conditions for the Green functions on the layers interface. The result will look exactly as (1) but with more complex  $G_{1,2}$ :

$$G_{1} = \frac{(s_{1} + s_{2})(p + s_{1})e^{\zeta_{n}s_{1}h/c} + (s_{1} - s_{2})(p - s_{1})e^{-\zeta_{n}s_{1}h/c}}{(s_{1} + s_{2})(p - s_{1})e^{\zeta_{n}s_{1}h/c} + (s_{1} - s_{2})(p + s_{1})e^{-\zeta_{n}s_{1}h/c}},$$

$$G_{2} = -\frac{\left(\varepsilon_{2}s_{1} + \varepsilon_{1}s_{2}\right)\left(\varepsilon_{1}p + s_{1}\right)e^{\zeta_{n}s_{1}h/c} + \left(\varepsilon_{2}s_{1} - \varepsilon_{1}s_{2}\right)\left(\varepsilon_{1}p - s_{1}\right)e^{-\zeta_{n}s_{1}h/c}}{\left(\varepsilon_{2}s_{1} + \varepsilon_{1}s_{2}\right)\left(\varepsilon_{1}p - s_{1}\right)e^{\zeta_{n}s_{1}h/c} + \left(\varepsilon_{2}s_{1} - \varepsilon_{1}s_{2}\right)\left(\varepsilon_{1}p + s_{1}\right)e^{-\zeta_{n}s_{1}h/c}},\qquad(5)$$

where  $s_{1,2}$  are defined similar to s in (2). The force between plate and sphere is given by (4) with the above  $G_{1,2}$ . Qualitatively the effect of the top layer will be negligible if  $h\omega_{1p}/c \ll 1$  where  $\omega_{1p}$  is the plasma frequency of this layer. For typical plasma frequencies  $\sim 10^{16} s^{-1}$  it is definitely not the case even for h = 8 nm. The force between layered bodies was found also in [13] with a little bit different technic but it was not used there for actual calculations.

Now we are able to evaluate the Casimir force in real geometry of the experiments if there is information on the dielectric functions of the used materials: Au, Al, and  $Au_{0.6}Pd_{0.4}$  alloy. Strictly speaking, one has to measure these functions in wide range of wavelengths on the samples which are used for the force measurement. It was not done in all experiments and to draw any conclusion from them we have to make some assumptions on  $\varepsilon(\omega)$ . At low frequencies Au and Al are well described by the Drude theory, where

$$\varepsilon\left(\omega\right) = 1 - \frac{\omega_p^2}{\omega\left(\omega + i\omega_\tau\right)}.\tag{6}$$

Here  $\omega_p$  is the free electron plasma frequency and  $\omega_{\tau}$  is the Drude damping frequency. A simple test for validity of (6) is the behavior of the material resistivity [14] that is defined as

$$\rho\left(\omega\right) = Im \frac{1}{\varepsilon_0 \left(1 - \varepsilon\left(\omega\right)\right) \omega} = \frac{\omega_\tau}{\varepsilon_0 \omega_p^2},\tag{7}$$

where  $\varepsilon_0$  is the free space permittivity. The resistivity is frequency independent within the Drude approximation. For crystalline samples of Au and Al (the entries 2 in Table 1) the frequency behavior of the resistivity and  $Im\varepsilon(\omega)$  are shown in Fig.1. The data on the dielectric functions were taken from [15], where the data from many original works are collected. Palladium definitely cannot be described by (6) at any frequency. However, it is known experimentally that amorphous metallic alloys such as Au/Pd can be described by the Drude approximation [14]. The physical explanation for this is associated with large Drude damping of the compounds like  $Au_{0.6}Pd_{0.4}$ . Eq.(7) allows to use well defined static resistivity  $\rho(0) = \rho_0$  instead of the damping frequency  $\omega_{\tau}$ .

Of course, at higher frequencies when interband transitions are reached the Drude approximation fails. Nevertheless, it is very useful since low frequencies dominate in the dispersion relation. Extrapolation of (6) to high frequencies gives

$$\varepsilon(i\zeta) = 1 + \frac{\omega_p^2}{\zeta(\zeta + \omega_\tau)}.$$
(8)

The relative error inserted in (8) due to extrapolation can be estimated as  $\omega_{\tau}/\omega_0$ , where  $\omega_0$  is the frequency of the first resonance for a given metal. The error can be as large as 10% but it does not influence significantly in the force. If we will use (8) for the force computation and change  $\omega_p$  by 5% (10% correction to  $\varepsilon(i\zeta)$  at all frequencies), then the force is changed less than 2%. Moreover, since the interband transitions give a correction to (8) which is frequency dependent, it reduces the correction to the force further below the experimental uncertainties. The possibility to neglect the interband transitions in Al for the force evaluation was noted in [8]. It agrees with our estimate and direct computation using the handbook data for  $Im\varepsilon(\omega)$ . Therefore, in all cases of interest we can use Eq.(8) to describe the dielectric function of a material on the imaginary axis. Since the integral in (3) is saturated in the low frequency region, we should extract the parameters  $\omega_p$  and  $\omega_{\tau}$ from the data for real and imaginary parts of  $\varepsilon(\omega)$  by fitting them in infrared region with (6).

It is important that optical properties of evaporated (spattered) films can be quite different from those of bulk material and depend on the technological details. It is known, for example, that the film density is typically 0.7 of that of the bulk material if it was not annealed. For the resistivity of spattered and evaporated Aufilms the value  $\rho_0 = 8.2 \ \mu\Omega \cdot cm$  has been reported [16] in contrast with the bulk

Al	$\omega_p \cdot 10^{-16}$	$\omega_{ au} \cdot 10^{-13}$	$\rho_0 \ \mu \Omega \cdot cm$
1*	$1.54\pm0.01$	$15.5\pm0.6$	7.39
2	$2.235\pm0.001$	$12.49\pm0.01$	2.83
3	$2.43\pm0.05$	$14.4\pm0.7$	2.76
4*	$1.63\pm0.03$	$18.2\pm0.7$	7.74
$\mathbf{A}\mathbf{u}$	$\omega_p \cdot 10^{-16}$	$\omega_{ au} \cdot 10^{-13}$	$\rho_0 \ \mu \Omega \cdot cm$
<b>Au</b> 1	$\frac{\omega_p \cdot 10^{-16}}{1.280 \pm 0.001}$	$\frac{\omega_{\tau} \cdot 10^{-13}}{3.29 \pm 0.05}$	$\frac{\rho_0 \ \mu \Omega \cdot cm}{2.27}$
Au           1           2	$\frac{\omega_p \cdot 10^{-16}}{1.280 \pm 0.001}$ $1.372 \pm 0.001$	$\frac{\omega_{\tau} \cdot 10^{-13}}{3.29 \pm 0.05}$ $4.060 \pm 0.002$	$\frac{\rho_0 \ \mu \Omega \cdot cm}{2.27}$ 2.44
Au           1           2           3	$ \frac{\omega_p \cdot 10^{-16}}{1.280 \pm 0.001} \\ 1.372 \pm 0.001 \\ 1.34 \pm 0.02 $	$ \frac{\omega_{\tau} \cdot 10^{-13}}{3.29 \pm 0.05} \\ 4.060 \pm 0.002 \\ 7.08 \pm 0.18 $	$ \begin{array}{c} \rho_0 \ \mu\Omega \cdot cm \\ 2.27 \\ 2.44 \\ 4.46 \end{array} $

Table 1: Drude parameters with statistical errors for Al and Au found by fitting  $\epsilon(\omega)$  in the infrared range ( $\lambda > 2 \ \mu m$ ). The stars in the first column mark the data for film samples.

resistivity 2.25  $\mu\Omega \cdot cm$ . All these make impossible to use the handbook data for reliable calculation of the Casimir force. This conclusion is illustrated by Table 1, where the parameters for Al and Au found by fitting the data from [15] are presented.

Though we cannot use handbook data to evaluate the force, one can confine it for a given experiment. This statement is based on the observation that because of better reflectivity the force (4) increases every time when  $\omega_p$  increases or  $\rho_0$ decreases. For us it is important that any technological procedures will reduce  $\omega_p$ and increase the resistivity  $\rho_0$ . The perfect crystalline material will have the largest plasma frequency and the smallest resistivity and these parameters are well defined. The plasma frequency  $\omega_p$  is defined by the concentration of free electrons in the metal n and their effective mass  $m_e^*$ 

$$\omega_p = \sqrt{\frac{e^2 n}{m_e^* \varepsilon_0}}.$$
(9)

Gold is a good conductor and  $m_e^*$  is quite close to the mass of electron. We will find the upper limit on the electron concentration if suppose that every Au atom produce a free electron. Then for Au plasma frequency one finds  $\omega_p^{Au} = 1.37 \cdot 10^{16} \ s^{-1}$ . The static resistivity can be used to get the damping frequency  $\omega_{\tau}$  with the help of (7) at a given  $\omega_p$ . For crystalline gold it is  $\rho_0^{Au} = 2.25 \ \mu\Omega \cdot cm$ . One can compare these parameters with that given in Table 1 to make sure that they correspond to the limit values. In the TP experiment [6] the bodies were covered with Au of thickness 0.5  $\mu m$  that is thick enough to be considered as infinite. Substituting the Au parameters into (8) and calculating the force according to (4) one finds the upper limit on the Casimir force in the TP experiment. The residual force  $F^{exp}(a_i) - F^{lim}(a_i)$  is shown in Fig.2a, where  $F^{exp}(a_i)$  are the experimental points at separations  $a_i$ . The prediction obviously does not contradict to the experiment but dealing with the upper limit we cannot conclude that there is an agreement, either.

For the AFM experiments [7, 8] the upper limit on the Casimir force is more restrictive. The plasma frequency for Al can be restricted using (9) if one supposes that every atom produces 3 free electrons. It gives  $\omega_p^{Al} = 2.40 \cdot 10^{16} \ s^{-1}$  that coincide with the largest value in Table 1. The resistivity of perfect crystal is  $\rho_0^{Al} = 2.65 \ \mu\Omega \cdot cm$ . Since we successfully predicted the plasma frequencies for the best samples of Au and Al, the same way one can use to estimate  $\omega_p$  for Au/Pd. If each Au atom gives one and Pd atom gives not more than two free electrons, then  $\omega_n^{Au/Pd} = 1.69 \cdot 10^{16} \ s^{-1}$ . This alloy is used in microelectronics and resistivity of the bulk material is known to be  $\rho_0^{Au/Pd} \approx 30 \ \mu\Omega \cdot cm$  [17] in accordance with the statement that alloys have large Drude damping. These data allow to find the upper limit on the force using (4) with the functions  $G_{1,2}$  defined in (5). Real surface of the bodies is always distorted. The distortion statistics were analysed with atomic force microscope [8, 11]. The force has to be averaged over the distorted surfaces and we use for this the procedure developed in [11]. This procedure seems quite reliable. Moreover, the important progress in controlled metal evaporation [8] allowed to reduce the surface roughness to the level when the correction to the force becomes practically unimportant for the experiment [8].

It was indicated [7] that the thickness of Au/Pd layer is less than 20 nm, that is why for calculations the conservative value h = 15 nm was chosen. The top layer changes the force on 13 pN at the smallest separation. Variation of  $\omega_p^{Al}$  on 10% gives only 1 pN change in the force because of screening effect of the top layer. The same variation in  $\omega_p^{Au/Pd}$  changes the force on 2 pN. The resistivity variation of the Au/Pd layer on 30% gives 1 pN effect. At larger separations all the effects become smaller. All this means that the limit is stable in respect to variation of the parameters. It is clear also that the top layer definitely cannot be ignored in the force evaluation. The residual force  $F^{exp}(a) - F^{lim}(a)$  with the experimental points from [7] is shown in Fig.2b by triangles.

In [8] the assumption of absolute transparency of the Au/Pd layer was not only used for theoretical interpretation of the result but also in the procedure of the force extraction from the raw data. For this reason we cannot use the points for the force directly. Fortunately, it is easy to restore the right data by shifting all the points to larger separations on 2h = 16 nm. The result for the residual force  $F^{exp}(a) - F^{lim}(a)$ with shifted experimental points from [8] is presented in Fig.2b by open squares. This figure clearly indicates the presence of some unexplained attractive force which is decreasing rapidly when the distance between bodies increased. One can speculate that the observed discrepancy is explained by a new Yukawa force mediated by a light scalar boson but we will not discuss now the restrictions on the Yukawa parameters that will be given elsewhere. To make the experiment absolutely clear, it is preferable to use Au instead of Al metallization because its non-reactive surface has strong advantage over Al. It excludes also additional uncertainties connected with Au/Pd layer. One can use as well silver or copper but they are not as inert as gold. In practice it is difficult to measure the dielectric function at the wavelengths larger than 30  $\mu m$  but this range gives an important contribution to the dispersion relation. That is why the material behavior in this range has to be predictable. One can say definitely that the materials of platinum group cannot be used since they are not described by (6) at low frequencies. An additional advantage of Au metallization is higher density of the bodies coating. In this case the hypotetic Yukawa force will be larger roughly in  $(\rho_{Au}/\rho_{Al})^2 \approx 50$  times. If the observed discrepancy has relation with the Yukawa interaction, the AFM experiment with Au metallization of the bodies will definitely reveal this new force even without detailed knowledge of optical properties of the metallization.

In conclusion, we have found the upper limit on the Casimir force that is realized for perfect crystalline coating of the bodies for which electrical and optical properties are well defined. This limit is smaller than the observed force in the AFM experiments and the difference far exceeds experimental errors and theoretical uncertainties for small separations between bodies. Au metallization of the bodies in the AFM experiment will allow to reveal origin of the discrepancy.

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## **Figure captions**

**Figure 1**. Validity of the Drude approximation for Al (triangles) and Au (circles) in the infrared range. The resistivity does not depend on frequency (left axis). Solid lines (right axis) demonstrate that  $Im\varepsilon(\omega)$  depends on  $\omega$  according to (6) with the parameters given in Table 1 (entries 2).

**Figure 2.** The residual force  $F^{exp}(a_i) - F^{lim}(a_i)$  for different experiments: (a) TP experiment [6]; (b) AFM experiments with the data from [7] (triangles) and from [8] (open squares).



