Stronger constraints on axion from measuring the Casimir interaction by means of dynamic atomic force microscope

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Abstract

We calculate the additional force due to two-axion exchange acting in a sphere-disc geometry, used in experiments on measuring the gradient of the Casimir force. With this result, stronger constraints on the pseudoscalar coupling constants of an axion and axion-like particles to a proton and a neutron are obtained over the wide range of axion masses from $3 \times 10^{-5}$ to 1 eV. Among the three experiments with Au-Au, Au-Ni and Ni-Ni boundary surfaces performed by means of dynamic atomic force microscope, major improving is achieved for the experiment with Au-Au test bodies. Here, the constraints obtained are stronger up to a factor of 170, as compared to the previously known ones. The largest strengthening holds for the axion mass 0.3 eV.

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I. INTRODUCTION

It happens that some predicted particle is of primary importance for theoretical reasons, but avoids experimental observation over many years. Commonly known predictions of this kind, which were finally confirmed, are the neutrino and the Higgs boson. In a similar way, the axion was predicted \cite{1, 2} in 1978 as a consequence of breaking of Peccei and Quinn symmetry \cite{3}, but has not been discovered up to the present. It is known that Peccei and Quinn symmetry provides the most natural possibility to avoid strong CP violation and the large electric dipole moment induced by it for the neutron in quantum chromodynamics. What is more, axion reasonably explains the nature of dark matter in astrophysics and cosmology \cite{4, 5}. Because of this, after the proper QCD axions were constrained to a narrow band in parameter space, different axion-like fields have been introduced, specifically, inspired by string theory \cite{6}. Some of them can be used to solve the problem of strong CP violation (see, for instance, the model of GUT axion \cite{7, 8} or various variants \cite{9, 10} of the model of hadronic axion \cite{11, 12}), but not all. Keeping in mind, however, that our approach in the search for axion and axion-like particles is common, below we use both terms more or less synonymously. There is an opinion that eventually the existence of the axion will be confirmed. For this reason, many experiments searching for the axion have been performed in the past and many more are planed for the near future.

It has been suggested \cite{13–15} to use the gravitational experiments of Eötvös \cite{15, 16} and Cavendish \cite{17} type for constraining the coupling constants of the pseudoscalar interaction of massless axions to protons and neutrons. This approach was generalized \cite{18} for the more realistic case of massive axions, and rather strong constraints were obtained in the mass range \( m_a \leq 9.9 \mu \text{eV} \). Constraints on the coupling constants of an axion to nucleons were also obtained from searching the Compton-like process for axions emitted from the Sun \cite{19} and from observations of the neutrino burst of supernova 1987A \cite{20}. Cosmology also places constraints on axion-like particles in different ranges of their mass if we take into account that axions could have been produced at the early stages of Universe evolution and contribute to the dark matter. Thus, thermal axions with \( m_a > 0.7 \text{eV} \) are excluded because they would make too large contribution to the hot dark matter \cite{21} with a reservation that such kind limits typically refer to specific couplings in some axion models and may be not applicable to all axion-like particles. At the same time, some models of cold dark matter...
exclude axions with $m_a < 10 \mu$eV \cite{19}.

The interaction with photons is used in many experimental searches for axions. Thus, strong constraints on the coupling constant of an axion to photon were obtained from searching axions emitted from the Sun by means of the CERN axion solar telescope \cite{22}. These constraints are relevant to axions with masses from $m_a = 0.39$ eV to $m_a = 0.64$ eV. Strong astrophysical constraints on the axion-photon interaction were found from gravitationally bound systems of stars of approximately the same age (globular clusters). These constraints are valid for axions with larger masses from about 0.3 eV to several tens keV \cite{19,23} (too heavy axions are also excluded due to their enormously large contribution to hot dark matter).

Besides nucleons and photons, axion may also interact with electrons. Interaction with electrons includes the Compton process and the electron-positron annihilation with emission of an axion in stellar plasmas. These processes are used for constraining the axion-electron coupling constant \cite{24,25}. As was mentioned above, experiments on the search of axions are numerous and varied (see reviews in Refs. \cite{6,26–28}). However, there is still the so-called window in the possible values of axion masses which extends \cite{19} from approximately $m_a = 10^{-5}$ eV to $m_a = 10^{-2}$ eV, where constraints on the axion parameters are rather weak (haloscope searches aim to narrow this window \cite{6}).

In Ref. \cite{29} strong constraints on the coupling constants of pseudoscalar interaction of axion to nucleons in the mass range from $10^{-4}$ eV to 0.3 eV were obtained from measurements of the thermal Casimir-Polder force between a Bose-Einstein condensate of $^{87}$Rb atoms and a SiO$_2$ plate \cite{30}. Experiments on measuring the Casimir force \cite{31} have long been used for obtaining constraints on the Yukawa-type corrections to Newton’s gravitational law (see review in Ref. \cite{32} and Refs. \cite{33–38} for the most recent results). The Yukawa-type correction terms to Newtonian gravity arise either due to exchange of light scalar particles between atoms of test bodies or from extra-dimensional physics with low-energy compactification scale \cite{39}. The axion is a pseudoscalar particle. As a result, the effective potential between two fermions, arising from the exchange of an axion with the pseudoscalar coupling to fermions, is spin-dependent \cite{18}. Keeping in mind that the test bodies in Casimir experiments are unpolarized, one concludes that there is no any extra force due to a single-axion exchange. In Ref. \cite{29} the additional spin-independent force between an atom and a plate was found as a result of two-axion exchange (in Ref. \cite{18} this process was used for
constraining the parameters of an axion from the gravitational experiments of Eötvos- and Cavendish-type).

In this paper, we obtain stronger constraints on the constants of pseudoscalar coupling of axion-like particles to a proton and a neutron which follow from experiments on measuring the gradient of the Casimir force between a sphere and a plate by means of dynamic atomic force microscope (AFM). We derive constraints following from the experiments with an Au-coated sphere and an Au-coated plate [40, 41], with an Au-coated sphere and a Ni-coated plate [42], and, finally, from the experiment with two Ni-coated test bodies [43, 44]. For this purpose, we calculate the gradient of the additional force arising between a sphere and a plate due to two-axion exchange taking into account the layer structure of both test bodies used in each experiment. The constraints obtained from each of the three experiments in the region of axion masses from $m_a = 2 \times 10^{-5}$ eV to $m_a = 1$ eV are compared between themselves and with those of Ref. [29]. It is shown that in the region of axion masses from $m_a = 10^{-4}$ eV to $m_a = 0.3$ eV, which was covered in Ref. [29], the constraints obtained in this work are significantly stronger. The largest strengthening by a factor of 170 holds for an axion mass $m_a = 0.3$ eV.

The paper is organized as follows. In Sec. II, we derive an expression for the gradient of the force arising between a sphere and a plate due to two-axion exchange. In Sec. III, this expression is adapted for the layer structure of the experiment with two Au-coated test bodies, and the respective constraints on the axion-nucleon coupling constants are obtained. In Sec. IV, the same is done for the experiment with an Au-coated sphere and a Ni-coated plate. Section V contains derivation of the constraints following from the experiment with two Ni-coated test bodies. In Sec. VI the reader will find our conclusions and discussion.

In what follows, the system of units with $\hbar = c = 1$ is used.

II. GRADIENT OF THE FORCE BETWEEN A SPHERE AND A PLATE DUE TO TWO-AXION EXCHANGE

In three experiments performed by means of AFM [40–44] the gradient of the Casimir force was measured between spheres of about 50 $\mu$m radius and discs of approximately 5 mm radius made of different materials. Taking into account that the characteristic size of the sphere is by a factor of 100 smaller than that of the disc, all points of the sphere can be
considered as situated above the disc center (i.e., the disc area can be considered as infinitely large). Here, we calculate the additional force acting in a sphere-plate configuration due to two-axion exchange between protons and neutrons belonging to these test bodies. In doing so, we assume the pseudoscalar character of an axion-fermion interaction and neglect the interaction of axions with electrons \[18\]. In any case, the account of possible scalar coupling of axions to fermions \[45\] or of axion-electron interaction could only slightly increase the force magnitude and, as a result, would lead to minor strengthening of the constraints obtained (see Refs. \[46, 47\] for the constraints on coupling constants of scalar interaction of axion with nucleons).

Let the coordinate plane \(x, y\) coincide with an upper plane surface of the disc of thickness \(D\) and let the \(z\) axis be perpendicular to it. We choose the origin of the coordinate system at the center of the upper surface of the disc. The sphere of radius \(R\) has its center at \(z = a + R\). The effective potential due to two-axion exchange between two nucleons (protons or neutrons) situated at the points \(r_2\) of the disc and \(r_1\) of the sphere is given by \[18, 48, 49\]

\[
V(|r_1 - r_2|) = -\frac{g_{ak}^2 g_{al}^2}{32\pi^3 m^2} \frac{m_a}{(r_1 - r_2)^2} K_1(2m_a|r_1 - r_2|).
\]  

(1)

Here, \(g_{ak}\) and \(g_{al}\) are the constants of a pseudoscalar axion-proton \((k, l = p)\) or axion-neutron \((k, l = n)\) interaction, \(m = (m_n + m_p)/2\) is the mean of the neutron and proton masses, and \(K_1(z)\) is the modified Bessel function of the second kind. In the derivation of Eq. (1) it was assumed that \(|r_1 - r_2| \gg 1/m\). This condition is fulfilled with large safety margin because in the experiments of Refs. \[40–44\] it holds \(a > 200\) nm.

Using Eq. (1), the additional force due to two-axion exchange, \(F_{\text{add}}\), can be obtained by integration over the volumes of the sphere \(V_s\) and of the disc \(V_d\)

\[
F_{\text{add}}(a) = -\int_{V_s} dr_1 \int_{V_d} dr_2 \frac{\partial}{\partial z_1} V(|r_1 - r_2|).
\]  

(2)

Now we introduce the polar coordinates on the disc and calculate the gradient of the force (2) taking into account Eq. (1)

\[
\frac{\partial F_{\text{add}}(a)}{\partial a} = \frac{\pi m_a}{m^2 m_H^2} C_d C_s \frac{\partial}{\partial a} \int_a^{2R+a} dz_1 [R^2 - (z_1 - R - a)^2] \times \frac{\partial}{\partial z_1} \int_{-D}^0 dz_2 \int_0^\infty \rho d\rho \frac{K_1(2m_a\sqrt{\rho^2 + (z_1 - z_2)^2})}{\rho^2 + (z_1 - z_2)^2}.
\]  

(3)

Here, the coefficient \(C_d\) for the disc material is defined as

\[
C_d = \rho_d \left( \frac{g_{ap}^2 Z_d}{4\pi \mu_d} + \frac{g_{an}^2 N_d}{4\pi \mu_d} \right),
\]  

(4)
where $\rho_d$ is the disc density, and $Z_d$ and $N_d$ are the number of protons and the mean number of neutrons in the disc atom (molecule). The quantity $\mu_d = m_d/m_H$, where $m_d$ and $m_H$ are the mean mass of the disc atom (molecule) and the mass of an atomic hydrogen, respectively. Note that the values of $Z/\mu$ and $N/\mu$ for the first 92 elements of the Periodic Table with account of their isotopic composition are presented in Ref. [50]. This information is used below in computations of Secs. III–V. The coefficient $C_s$ for a sphere material is defined similarly to Eq. (4) with a replacement of all indices $d$ for $s$.

It is convenient to calculate first the quantity

$$I \equiv \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 \int_{0}^{\infty} \rho dp \frac{K_1(2m_a \sqrt{\rho^2 + (z_1 - z_2)^2})}{\rho^2 + (z_1 - z_2)^2}$$

entering Eq. (3). Using the integral representation [51]

$$\frac{K_1(z)}{z} = \int_{1}^{\infty} du \sqrt{u^2 - 1} e^{-zu},$$

one can rearrange Eq. (5) in the form

$$I = 2m_a \int_{1}^{\infty} du \sqrt{u^2 - 1} \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 e^{-2m_a u (z_1 - z_2)}.$$

Now we introduce the new variable $v = \sqrt{\rho^2 + (z_1 - z_2)^2}$ instead of $\rho$ in Eq. (7) and integrate with respect to it

$$I = \int_{1}^{\infty} du \sqrt{u^2 - 1} \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 e^{-2m_a u (z_1 - z_2)}.$$

Integrating and differentiating in Eq. (8) with respect to $z_2$ and $z_1$, respectively, one obtains

$$I = -\int_{1}^{\infty} du \sqrt{u^2 - 1} \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 e^{-2m_a u (z_1 - z_2)}.$$

Substituting Eq. (9) in Eq. (3) and differentiating with respect to $a$, we arrive at

$$\frac{\partial F_{\text{add}}(a)}{\partial a} = \frac{2\pi m_a}{m^2 m_H^2} C_d C_s \int_{1}^{\infty} du \sqrt{u^2 - 1} \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 e^{-2m_a u (z_1 - z_2)}.$$

Finally, calculating the integral with respect to $z_1$, we find

$$\frac{\partial F_{\text{add}}(a)}{\partial a} = \frac{\pi}{m^2 m_H^2} C_d C_s \int_{1}^{\infty} du \sqrt{u^2 - 1} \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 e^{-2m_a u (z_1 - z_2)}.$$

$$= \frac{2\pi m_a}{m^2 m_H^2} C_d C_s \int_{1}^{\infty} du \sqrt{u^2 - 1} \frac{\partial}{\partial z_1} \int_{-D}^{0} dz_2 e^{-2m_a u (z_1 - z_2)}.$$
where the following notation is introduced

\[
\Phi(r, z) = r - \frac{1}{2z} + e^{-2rz} \left( r + \frac{1}{2z} \right).
\]  

(12)

From Eq. (11) one can simply obtain the additional force gradient due to two-axion exchange between a spherical envelope of radius \(R\) and thickness \(\Delta_s\) and a plate. This can be found by subtracting from Eq. (11) the force due to a sphere of radius \(R - \Delta_s\) placed at a separation \(a + \Delta_s\) from the plate:

\[
\frac{\partial F_{\text{add}}(a)}{\partial a} = \frac{\pi}{m^2 m_H^2 C_d C_s} \int_1^{\infty} du \frac{\sqrt{u^2 - 1}}{u^2} \left( 1 - e^{-2m_a u D} \right) \times e^{-2m_a u} \left[ \Phi(R, m_a u) - e^{-2m_a \Delta_s} \Phi(R - \Delta_s, m_a u) \right].
\]  

(13)

In the end of this section, we note that the relative contribution of the boundary effects arising due to a finiteness of the plate area to the force gradient (13) is of the order

\[
\delta \left( \frac{\partial F_{\text{add}}}{\partial a} \right) \sim \frac{R}{R_d m_a},
\]  

(14)

where \(R_d\) is the disc radius. Taking into account the values of the experimental parameters indicated above, one can conclude that the contribution of the boundary effects in the additional force gradient (13) does not exceed 1% for axion masses satisfying the condition \(m_a > 1/R_d \approx 4 \times 10^{-5}\) eV.

III. CONSTRAINTS FROM EXPERIMENT WITH TWO GOLD-COATED TEST BODIES

In the experiment of Refs. [40, 41], the dynamic AFM was used to measure the gradient of the Casimir force between an Au-coated hollow sphere (spherical envelope) of \(R = 41.3\) \(\mu\)m radius made of fused silica and an Au-coated thick sapphire disc of \(R_d = 5\) mm radius. The thickness of glass spherical envelope was \(\Delta_{gs} = 5\) \(\mu\)m. The thicknesses and density of the Au coatings on the sphere and the disc were \(\Delta_{gs}^\text{Au} = \Delta_{d}^\text{Au} = 280\) nm and \(\rho_{\text{Au}} = 19.29\) g/cm\(^3\), respectively. In relation to the Casimir force this thickness is quite sufficient for considering the sphere and the disc as all-gold bodies [32]. However, the additional force due to two-axion exchange is highly sensitive to the material composition of both test bodies, and this should be taken into account in computations. In the experiment of Refs. [40, 41], the Au layers on both test bodies were deposited on the layer of Al of thickness \(\Delta_{gs}^\text{Al} = \Delta_{d}^\text{Al} = 20\) nm.
with density \( \rho_{\text{Al}} = 2.7 \text{ g/cm}^3 \). The density of glass (SiO\(_2\), the material of the spherical envelope) was \( \rho_g = 2.5 \text{ g/cm}^3 \), whereas the density of sapphire (Al\(_2\)O\(_3\), the material of the disc) was \( \rho_{\text{sa}} = 4.1 \text{ g/cm}^3 \).

By applying Eq. (13) to each pair of material layers forming the spherical envelope and the disc, we arrive at the following expression for the gradient of the additional force arising from the two-axion exchange:

\[
\frac{\partial F_{\text{add}}(a)}{\partial a} = \frac{\pi}{m_2^2 m_2^2} \int_1^{\infty} du \frac{\sqrt{u^2 - 1}}{u^2} e^{-2m_2au} X_s(m_a u) X_d(m_a u),
\]

(15)

where the functions \( X_d \) and \( X_s \) are defined as follows:

\[
X_d(z) \equiv C_{\text{Au}} \left( 1 - e^{-2z\Delta_{\text{Au}}} \right) + C_{\text{Al}} e^{-2z\Delta_{\text{Al}}} \left( 1 - e^{-2z\Delta_{\text{Al}}} \right) + C_{\text{sa}} e^{-2z(\Delta_{\text{Au}} + \Delta_{\text{Al}})}
\]

\[
X_s(z) \equiv C_{\text{Au}} \left[ \Phi(R, z) - e^{-2z\Delta_{\text{Au}}} \Phi(R - \Delta_{\text{Au}}, z) \right]
+ C_{\text{Al}} e^{-2z\Delta_{\text{Al}}} \left[ \Phi(R - \Delta_{\text{Al}}, z) - e^{-2z\Delta_{\text{Al}}} \Phi(R - \Delta_{\text{Al}} - \Delta_{\text{Al}}, z) \right]
+ C_{\text{g}} e^{-2z(\Delta_{\text{Au}} + \Delta_{\text{Al}})} \left[ \Phi(R - \Delta_{\text{Au}} - \Delta_{\text{Al}}, z) - e^{-2z\Delta_{\text{Al}}} \Phi(R - \Delta_{\text{Au}} - \Delta_{\text{Al}} - \Delta_{\text{g}}, z) \right].
\]

(16)

Note that the thickness of the sapphire disc is put equal to infinity, as it does not influence the result. The coefficients \( C_{\text{Au}}, C_{\text{Al}}, C_{\text{sa}} \) and \( C_{\text{g}} \) are defined in Eq. (4), as applied to the atoms Au, Al and to the molecules Al\(_2\)O\(_3\), SiO\(_2\), respectively. In Ref. [50] one finds the values of the following multiples entering Eq. (4):

\[
\frac{Z_{\text{Au}}}{\mu_{\text{Au}}} = 0.40422, \quad \frac{N_{\text{Au}}}{\mu_{\text{Au}}} = 0.60378,
\]

(17)

\[
\frac{Z_{\text{Al}}}{\mu_{\text{Al}}} = 0.48558, \quad \frac{N_{\text{Al}}}{\mu_{\text{Al}}} = 0.52304.
\]

For a sapphire molecule Al\(_2\)O\(_3\), in accordance to Ref. [50], we have

\[
\frac{Z_{\text{Al}_{2}\text{O}_3}}{\mu_{\text{Al}_{2}\text{O}_3}} = \frac{2Z_{\text{Al}} + 3Z_{\text{O}}}{2\mu_{\text{Al}} + 3\mu_{\text{O}}} = 0.49422,
\]

(18)

\[
\frac{N_{\text{Al}_{2}\text{O}_3}}{\mu_{\text{Al}_{2}\text{O}_3}} = \frac{2N_{\text{Al}} + 3N_{\text{O}}}{2\mu_{\text{Al}} + 3\mu_{\text{O}}} = 0.51412.
\]

In a similar way, for fused silica molecule SiO\(_2\) we obtain [29, 50]

\[
\frac{Z_{\text{SiO}_2}}{\mu_{\text{SiO}_2}} = 0.503205, \quad \frac{N_{\text{SiO}_2}}{\mu_{\text{SiO}_2}} = 0.505179.
\]

(19)
The experimental data of Refs. [40, 41] for the gradient of the Casimir force $F'_C(a)$ were measured at separation distances $a \geq 235$ nm and were found to be in agreement with theoretical predictions of the Lifshitz theory of dispersion forces [32, 52] under the condition that the low-frequency behavior of the dielectric permittivity of Au is described by the plasma model. This means that the gradient of any additional force due to two-axion exchange is constrained by the inequality

$$\frac{\partial F_{\text{add}}(a)}{\partial a} \leq \Delta F'_C(a).$$

(20)

Here, $\Delta F'_C(a)$ is the total absolute error in the measured gradient of the Casimir force, which was determined in Refs. [40, 41] as a combination of random and systematic errors at a 67% confidence level.

We have substituted Eq. (15) in Eq. (20) and found numerically the regions of the axion parameters, $g_{ap}$, $g_{an}$ and $m_a$, where the inequality (20) is satisfied. The most strong constraints were obtained at the shortest experimental separation $a = 235$ nm, where the absolute error $\Delta F'_C = 0.5 \mu\text{N/m}$ [40, 41]. Note that the gradient of the Casimir force at this separation is equal to $F'_C = 73.86 \mu\text{N/m}$. In Fig. 1, we present the obtained constraints on the pseudoscalar coupling constants $g_{ap(n)}^2/(4\pi)$ as a function of the axion mass $m_a$. The solid lines from bottom to top are plotted under the conditions $g_{ap}^2 = g_{an}^2$, $g_{an}^2 \gg g_{ap}^2$ and $g_{ap}^2 \gg g_{an}^2$, respectively. The regions above each line are prohibited by the measurement data of Refs. [40, 41], and the regions below each line are allowed by the data. In Table I, the maximum allowed values of the pseudoscalar coupling constants of an axion-nucleon interaction for different values of the axion mass (column 1) are listed under the condition $g_{ap}^2 = g_{an}^2$ (column 2), $g_{an}^2 \gg g_{ap}^2$ (column 3), and $g_{ap}^2 \gg g_{an}^2$ (column 4). As can be seen in Fig. 1, for $m_a \leq 0.001$ eV the obtained constraints are almost independent of the axion mass (we do not extend our constraints to smaller $m_a$ where the omitted boundary effects may exceed 1%).

In Fig. 2, we compare the constraints of Fig. 1 and Table I, found under the most reasonable condition $g_{ap}^2 = g_{an}^2$, with the constraints of Ref. [29] obtained from measurements of the thermal Casimir-Polder force between the Bose-Einstein condensate of $^{87}$Rb atoms and SiO$_2$ plate [30]. The solid line reproduces the lowest line of Fig. 1 within an interval $10^{-4}$ eV $\leq m_a \leq 0.3$ eV, where the constraints of Ref. [29] were obtained, and the dashed line shows the respective constraints of Ref. [29]. As can be seen in Fig. 2, the experiment
on measuring the gradient of the Casimir force by means of dynamic AFM \[40, 41\] leads to stronger constraints than the experiment of Ref. \[30\] over the entire mass range of the latter. The largest strengthening by a factor of 170 holds for the axion mass \(m_a = 0.3\) eV. In the region of small axion masses \(m_a \lesssim 0.003\) eV, the strengthening is by a factor of 1.2. Similar results are obtained under alternative assumptions about the relationship between \(g_{ap}\) and \(g_{an}\). Thus, if \(g_{an} \gg g_{ap}\), the constraints of Fig. 1 are stronger than those of Ref. \[29\] up to a factor 185. The largest strengthening is achieved at the same axion mass \(m_a = 0.3\) eV. Under the assumption \(g_{ap} \gg g_{an}\), the respective strengthening is up to a factor 150 and again holds at \(m_a = 0.3\) eV. These results demonstrate that measurements of the gradient of the Casimir force by means of dynamic AFM should be considered as a useful supplementary method in the search for an axion.

**IV. CONSTRAINTS FROM EXPERIMENT WITH GOLD-COATED SPHERE AND NICKEL-COATED PLATE**

In Ref. [42], the dynamic AFM was used to measure the gradient of the Casimir force between an Au-coated hollow sphere of \(R = 64.1\) \(\mu\)m radius made of fused silica and a Ni-coated Si disc of density \(\rho_{Si} = 2.33\) g/cm\(^3\), which, again, can be considered as infinitely large. The thickness of glass spherical envelope was \(\Delta_{gs} = 5\) \(\mu\)m, i.e., the same as in the experiment of Refs. [40, 41]. It was coated by the layers of Al and Au of the same thicknesses \(\Delta_{s}^{Al}\) and \(\Delta_{s}^{Au}\), as in the experiment with two Au surfaces. The Si disc was coated with a single layer of magnetic metal Ni having the thickness \(\Delta_{d}^{Ni} = 154\) nm and density \(\rho_{Ni} = 8.9\) g/cm\(^3\).

The gradient of the additional force between the hollow sphere and the plate due to two-axion exchange in this experiment is, again, given by Eq. (15), where the function \(X_s\) is presented in Eq. (16). As to the function \(X_d\), it takes a more simple form than in Eq. (16), due to the presence of only one coating layer on the disc surface. For this function one obtains

\[
X_d(z) = C_{Ni} \left( 1 - e^{-2z\Delta_{d}^{Ni}} \right) + C_{Si} e^{-2z\Delta_{d}^{Ni}}. \tag{21}
\]

The coefficients \(C_{Ni}\) and \(C_{Si}\) are defined by Eq. (4) where the values \(Z/\mu\) and \(N/\mu\) are
given by \(50\)

\[
\begin{align*}
\frac{Z_{\text{Ni}}}{\mu_{\text{Ni}}} &= 0.48069, & \frac{N_{\text{Ni}}}{\mu_{\text{Ni}}} &= 0.52827, \\
\frac{Z_{\text{Si}}}{\mu_{\text{Si}}} &= 0.50238, & \frac{N_{\text{Si}}}{\mu_{\text{Si}}} &= 0.50628.
\end{align*}
\] (22)

The experimental data of Ref. [42] for \(F'_C(a)\) were taken at separation distances \(a \geq 220\) nm and found in agreement with the Lifshitz theory \[32, 52\] generalized for the case of magnetic materials. Specific characteristic feature of the experiment with Au-Ni test bodies, as compared with the case of two Au bodies considered in Sec. III, is that here the theoretical prediction over the entire measurement range does not depend on whether the low-frequency behavior of the dielectric permittivity of metals is described by the plasma model or by the Drude model. This makes the theory-experiment comparison fully transparent and independent of contribution of any background effect (a discussion on the Drude and plasma model approaches in the theory of dispersion forces can be found in Ref. [31]).

Taking into account that in the limits of the total experimental error, \(\Delta F'_C(a)\), no additional force was observed, the constraints on the parameters of the axion can be again derived from Eq. (20), where the gradient of the force due to two-axion exchange is given by Eq. (15) with the functions \(X_s\) and \(X_d\) defined in Eqs. (16) and (21), respectively.

The numerical analysis of the inequality (20) shows that the strongest constraints are obtained at the shortest experimental separation \(a = 220\) nm, where the total experimental error determined at a 67% confidence level is equal to \(\Delta F'_C = 0.79\, \mu N/m\) [42]. This should be compared with the value of the Casimir force gradient \(F'_C = 131.03\, \mu N/m\) measured at this separation. In Fig. 3, the solid lines show the constraints on the pseudoscalar coupling constants \(g^2_{ap(n)}/(4\pi)\), as functions of the axion mass \(m_a\), which follow from the experiment [42] with Au-Ni test bodies. The lines from bottom to top are plotted under the conditions \(g^2_{ap} = g^2_{an}, g^2_{an} \gg g^2_{ap}\), and \(g^2_{ap} \gg g^2_{an}\), respectively. As in Fig. 1, the regions above each line are prohibited by the results of experiment [42], and the regions below each line are allowed.

For comparison purposes, in the same figure we reproduce by the dashed lines the constraints of Fig. 1 obtained in Sec. III under the same respective conditions from the experiment with two Au surfaces. As can be seen in Fig. 3, the constraints following from the experiment with Au-Ni test bodies [42] are up to a factor 1.5 weaker than the constraints from the experiment with Au-Au bodies [40, 41]. This is mostly determined by the smaller density of Ni, as compared with Au. At the same time, the constraints shown by the solid
and dashed lines demonstrate the same behavior as functions of $m_a$. Taking into account that the experiment with Au-Ni test bodies is in a very good agreement with the Lifshitz theory with no additional assumptions concerning the low-frequency behavior of the dielectric permittivities, this provides greater confidence in the constraints of Sec. III obtained from the experiment of Refs. [40, 41] with two Au test bodies.

V. CONSTRAINTS FROM EXPERIMENT WITH TWO NICKEL TEST BODIES

In Refs. [43, 44] the gradient of the Casimir force was measured between a Ni-coated hollow glass sphere of $R = 61.71 \mu m$ radius and $\Delta_g = 5 \mu m$ thickness and a Ni-coated Si disc. As in the experiments of Refs. [40–42], the measurements were performed by means of dynamic AFM. For technological purposes, there were, however, two additional coatings on both test bodies in the experiment of Refs. [43, 44]. The outer layers of Ni were of thicknesses $\Delta_{Ni}^s = 210$ nm and $\Delta_{Ni}^d = 250$ nm on the spherical envelope and on the disc, respectively. The thicknesses of Al-coatings on both test bodies were equal $\Delta_{Al}^s = \Delta_{Al}^d = 40$ nm. There were also additional thin layers of Cr below Al layers with thicknesses $\Delta_{Cr}^s = \Delta_{Cr}^d = 10$ nm. This layer structure should be taken into account in the calculation of the additional force due to two-axion exchange.

After application of Eq. (13) to each pair of material layers forming the hollow sphere and the disc, one obtains Eq. (15) for the gradient of the additional force. In this case, however, due to a more complicated layer structure, the functions $X_d$ and $X_s$ take the form

$$X_d(z) = C_{Ni}\left(1 - e^{-2z\Delta_{Ni}^d}\right) + C_{Al}e^{-2z\Delta_{Ni}^d}\left(1 - e^{-2z\Delta_{Al}^d}\right) + C_{Cr}e^{-2z(\Delta_{Ni}^d+\Delta_{Al}^d)}\left(1 - e^{-2z\Delta_{Cr}^d}\right) + C_{Si}e^{-2z(\Delta_{Ni}^d+\Delta_{Al}^d+\Delta_{Cr}^d)},$$

$$X_s(z) = C_{Ni}\left[\Phi(R, z) - e^{-2z\Delta_{Ni}^s}\Phi(R - \Delta_{Ni}^s, z)\right] + C_{Al}e^{-2z\Delta_{Ni}^s}\left[\Phi(R - \Delta_{Ni}^s, z) - e^{-2z\Delta_{Al}^s}\Phi(R - \Delta_{Ni}^s - \Delta_{Al}^s, z)\right] + C_{Cr}e^{-2z(\Delta_{Ni}^s+\Delta_{Al}^s)}\left[\Phi(R - \Delta_{Ni}^s - \Delta_{Al}^s, z) - e^{-2z\Delta_{Cr}^s}\Phi(R - \Delta_{Ni}^s - \Delta_{Al}^s - \Delta_{Cr}^s, z)\right] + C_{g}e^{-2z(\Delta_{Ni}^s+\Delta_{Al}^s+\Delta_{Cr}^s)}\left[\Phi(R - \Delta_{Ni}^s - \Delta_{Al}^s - \Delta_{Cr}^s, z) - e^{-2z\Delta_{g}^s}\Phi(R - \Delta_{Ni}^s - \Delta_{Al}^s - \Delta_{Cr}^s - \Delta_{g}^s, z)\right],$$

where the function $\Phi(r, z)$ is defined in Eq. (12).
The coefficient $C_{Cr}$, which was not used above, is given by Eq. (4), where the values of $Z/\mu$ and $N/\mu$ for Cr can be found in Ref. [50],

$$\frac{Z_{Cr}}{\mu_{Cr}} = 0.46518, \quad \frac{N_{Cr}}{\mu_{Cr}} = 0.54379,$$

(24)

and the density of Cr is $\rho_{Cr} = 7.14 \text{ g/cm}^3$.

Measurements of the gradient of the Casimir force $F'_C$ in Refs. [43, 44] were performed at separations $a \geq 223$ nm. The experimental data were found in agreement with theoretical predictions of the Lifshitz theory [32, 52], generalized for the case of magnetic materials, under the condition that the low-frequency behavior of the dielectric permittivity of Ni is described by the plasma model [43, 44]. The predictions of the Lifshitz theory using the low-frequency behavior described by the Drude model was excluded by the data. The same result concerning the plasma and Drude models was obtained from the experiment using two Au test bodies discussed in Sec. III and from three independent measurements of Refs. [53–56] performed with two Au test bodies using an alternative experimental technique (a micromachined oscillator). The important characteristic feature of the case of two magnetic surfaces is that here $F'_{C,D} > F'_{C,p}$, where the indices $D$ and $p$ denote which model, the Drude or the plasma, is used in computations for the dielectric permittivity at low frequencies. This is just the opposite to the case of two nonmagnetic surfaces, Au for instance, where $F'_{C,D} < F'_{C,p}$ [43, 44]. Taking into account that the experiments of Refs. [40, 41] with two Au surfaces, on the one hand, and of Refs. [43, 44] with two Ni surfaces, on the other hand, were performed using one and the same setup (the dynamic AFM) and in both cases the predictions of the Lifshitz theory $F'_{C,p}$ was confirmed, one can reliably exclude the role of any unaccounted systematic effect in the theory-experiment comparison. Note that the experiment [57], claiming confirmation of the Lifshitz theory combined with the Drude model, is not a direct measurement of the Casimir force, but up to an order of magnitude larger force of unknown origin. At separations below 3 \mu m the results of this experiment are in fact uncertain because it does not take into account glass imperfections, which unavoidably present on lenses of large radii [58]. At larger separations the measurement data of Ref. [57] better agrees with the plasma model [59].

We also emphasize that the difference $F'_{C,p} - F'_{C,D}$ cannot be approximated either by the interaction due to two-axion exchange or by the hypothetical Yukawa-type interaction [33–38]. Moreover, even if any other compensating interaction due to some unknown agent
existed and, being added to $F'_{C,D}$, brought the measurement data in agreement with $F'_{C,p}$ for Au-Au test bodies, it would only increase the deviations between experiment and theory for Ni-Ni test bodies and vice versa. The reason is that for Au-Au bodies this difference corresponds to the attractive force and for Ni-Ni to a repulsive. Thus, the disagreement of the data with $F'_{C,D}$ cannot be attributed to the influence of any additional force.

On this basis, we can obtain the constraints on the parameters of an axion from Eq. (20) expressing the measure of agreement of the measured data with $F'_{C,p}$. The strongest constraints follow at the shortest experimental separation $a = 223\, \text{nm}$, where the total experimental error determined at a 67% confidence level is equal to $\Delta F'_{C} = 1.2 \, \mu \text{N/m}^4$ [43, 44]. This should be compared with the gradient of the Casimir force measured at the shortest separation $F'_C = 131.03 \, \mu \text{N/m}$. In Fig. 4 we show the obtained constraints on the coupling constants $g_{ap(n)}^2/(4\pi)$ by the solid lines, as functions of the axion mass $m_a$. The lines from bottom to top are plotted under the conditions $g_{an}^2 = g_{ap}^2$, $g_{an}^2 \gg g_{ap}^2$ and $g_{ap}^2 \gg g_{an}^2$, respectively. The regions above each line are prohibited and below each line are allowed by the results of experiment of Refs. [43, 44]. As can be seen in Fig. 4, the two upper lines, calculated under conditions $g_{an}^2 \gg g_{ap}^2$ and $g_{ap}^2 \gg g_{an}^2$, almost overlap. This is caused by the fact that for Ni the difference of the quantities $Z/\mu$ and $N/\mu$ is much smaller than for Au.

In Fig. 4, we also reproduce by the dashed lines the constraints of Fig. 1 obtained under the same relationships between the coupling constants from the experiment with two Au test bodies [40, 41]. As is seen in Fig. 4, the experiment with two Ni test bodies leads to weaker constraints than the experiment with Au-Au bodies. The qualitative character of the dashed lines in Fig. 4 is, however, the same as of the solid lines, and weaker constraints obtained are explained by the smaller density of Ni, as compared with Au. As noted above, the experiment with two Ni surfaces provides further evidence for the absence of unaccounted systematic effects in measurements by means of dynamic AFM. Thus, the strongest constraints on the coupling constants of an axion to nucleons obtained using this setup (see Fig. 1 and Table I) can be considered as the best laboratory limits within the region of axion masses from about $10^{-4}$ eV to 1 eV.
VI. CONCLUSIONS AND DISCUSSION

In this paper, we have derived the constraints on the pseudoscalar coupling constants of axion-like particles to a proton and a neutron which follow from measurements of the gradient of the Casimir force in a sphere-plane geometry by means of dynamic AFM. For this purpose, three recent experiments have been analyzed: with an Au-coated sphere and an Au-coated disc [40, 41], with an Au-coated sphere and a Ni-coated disc [42], and with two Ni-coated test bodies [43, 44]. Taking into consideration that the test bodies in measurements of the Casimir interaction are unpolarized, we have calculated the gradient of the additional force arising between them due to two-axion exchange. This was done taking into account the layer structure of both test bodies specific for each experiment.

In all three experiments considered the measurement data for the gradient of the Casimir force were found in good agreement with theoretical predictions of the Lifshitz theory of dispersion forces, and no additional interaction was observed. This allowed to constrain the gradient of the additional force by the total experimental error in measurements of the gradient of the Casimir force. It was shown that the strongest constraints on the parameters of an axion follow from the experiment with two Au test bodies. However, two other experiments using magnetic (Ni) surfaces provide further support to these constraints by confirming the absence of any unaccounted systematic effect in the measurements. The obtained constraints are relevant to the wide region of axion masses from $3 \times 10^{-5}$ to 1 eV and are determined at the 67% confidence level. Using specific axion models, they can be conversed into limits on the angle $\theta_{\text{eff}}$ which measures CP violating effects. For this purpose, however, one would need to use respective constraints on a product of the pseudoscalar and scalar axion-nucleon coupling constants [60].

We have compared the derived constraints with the previously known laboratory constraints of Ref. [29] following from the measurements of thermal Casimir-Polder force [30]. It was found that the constraints obtained in this paper are stronger over the entire range of the axion mass. Under the assumption that $g_{an} = g_{ap}$, the largest strengthening by the factor of 170 is achieved for the axion mass $m_a = 0.3 \text{eV}$. Note that many constraints on axions with $m_a \lesssim 1 \text{eV}$ using interactions with photons, electrons and nucleons are based on the data of CERN axion solar telescope [22], globular cluster stars [19, 23], and the duration of neutrino burst of supernova 1987A [20]. These constraints are obtained from
astrophysics and, broadly speaking, may be more model-dependent, as compared to table-top laboratory experiments on measuring the Casimir interaction. In this respect the pure-laboratory experiment which searches for photon oscillations into weakly interacting sub-eV particles is of much interest. It is important also that our constraints are most strong in the region of axion masses $4 \times 10^{-5} \text{eV} < m_a < 0.3 \text{eV}$ partially overlapping with the axion window where model-independent constraints on the axion parameters are rather weak.

One can conclude that laboratory experiments on measuring the Casimir interaction have considerable opportunity in the search for an axion and further constraining its parameters.

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FIG. 1: (Color online) Constraints on the coupling constants of an axion to a proton or a neutron obtained from measurements of the gradient of the Casimir force between Au surfaces as functions of the axion mass. The lines from bottom to top are plotted under the conditions $g_{ap}^2 = g_{an}^2$, $g_{an}^2 \gg g_{ap}^2$, and $g_{ap}^2 \gg g_{an}^2$, respectively. The regions above each line are prohibited and below each line are allowed.
FIG. 2: (Color online) Constraints on the coupling constants of an axion to a nucleon under the condition $g_{ap}^2 = g_{an}^2$ as functions of the axion mass. The solid and dashed lines show the results obtained in this paper from measurements of the gradient of the Casimir force between Au surfaces [40, 41] and in Ref. [29] from measurements of the thermal Casimir-Polder force [30], respectively. The regions of the plane above each line are prohibited and below each line are allowed.
FIG. 3: (Color online) Constraints on the coupling constants of an axion to a proton or a neutron following from the experiment with Au-Ni test bodies (solid lines) and Au-Au test bodies (dashed lines) are shown as functions of the axion mass. The lines from bottom to top are plotted under the conditions $g_{ap}^2 = g_{an}^2$, $g_{an}^2 \gg g_{ap}^2$, and $g_{ap}^2 \gg g_{an}^2$, respectively. The regions above each line are prohibited and below each line are allowed.
FIG. 4: (Color online) Constraints on the coupling constants of an axion to a proton or a neutron following from the experiment with Ni-Ni test bodies (solid lines) and Au-Au test bodies (dashed lines) are shown as functions of the axion mass. The lines from bottom to top are plotted under the conditions $g_{ap}^2 = g_{an}^2$, $g_{an}^2 \gg g_{ap}^2$, and $g_{ap}^2 \gg g_{an}^2$, respectively. The regions above each line are prohibited and below each line are allowed.
TABLE I: Maximum values of the coupling constants of an axion to a proton and a neutron, allowed by measurements of the gradient of the Casimir force between Au surfaces, are calculated for different axion masses (column 1) under the conditions $g_{ap}^2 = g_{an}^2$ (column 2), $g_{an}^2 \gg g_{ap}^2$ (column 3), and $g_{ap}^2 \gg g_{an}^2$ (column 4).

| $m_a$ (eV) | $g_{ap}^2 = g_{an}^2$ | $g_{an}^2 \gg g_{ap}^2$ | $g_{ap}^2 \gg g_{an}^2$ |
|------------|-----------------------|-----------------------|-----------------------|
| \leq 0.001 | 1.92 x 10^{-4}        | 3.32 x 10^{-4}        | 4.54 x 10^{-4}        |
| 0.01       | 1.97 x 10^{-4}        | 3.39 x 10^{-4}        | 4.69 x 10^{-4}        |
| 0.05       | 2.15 x 10^{-4}        | 3.66 x 10^{-4}        | 5.18 x 10^{-4}        |
| 0.1        | 2.36 x 10^{-4}        | 4.00 x 10^{-4}        | 5.74 x 10^{-4}        |
| 0.2        | 2.83 x 10^{-4}        | 4.76 x 10^{-4}        | 6.95 x 10^{-4}        |
| 0.3        | 3.38 x 10^{-4}        | 5.69 x 10^{-4}        | 8.36 x 10^{-4}        |
| 0.4        | 4.05 x 10^{-4}        | 6.79 x 10^{-4}        | 1.00 x 10^{-3}        |
| 0.5        | 4.84 x 10^{-4}        | 8.10 x 10^{-4}        | 1.20 x 10^{-3}        |
| 0.6        | 5.77 x 10^{-4}        | 9.65 x 10^{-4}        | 1.43 x 10^{-3}        |
| 0.7        | 6.86 x 10^{-4}        | 1.15 x 10^{-3}        | 1.71 x 10^{-3}        |
| 0.8        | 8.15 x 10^{-4}        | 1.36 x 10^{-3}        | 2.03 x 10^{-3}        |
| 0.9        | 9.65 x 10^{-4}        | 1.61 x 10^{-3}        | 2.40 x 10^{-3}        |
| 1.0        | 1.14 x 10^{-3}        | 1.90 x 10^{-3}        | 2.84 x 10^{-3}        |