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Particles with nonlinear electric response: Suppressing van der Waals forces by an external field

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We study the classical thermal component of Casimir, or van der Waals, forces between point particles with highly anharmonic dipole Hamiltonians when they are subjected to an external electric field. Using a model for which the individual dipole moments saturate in a strong field (a model that mimics the charges in a neutral, perfectly conducting sphere), we find that the resulting Casimir force depends strongly on the strength of the field, as demonstrated by analytical results. For a certain angle between the external field and center-to-center axis, the fluctuation force can be tuned and suppressed to arbitrarily small values. We compare the forces between these particles with those between particles with harmonic Hamiltonians and also provide a simple formula for asymptotically large external fields, which we expect to be generally valid for the case of saturating dipole moments.

I. INTRODUCTION

Neutral bodies exhibit attractive forces, called van der Waals or Casimir forces depending on context. The earliest calculations were formulated by Casimir, who studied the force between two metallic parallel plates [1], and generalized by Lifshitz [2] for the case of dielectric materials. Casimir and Polder found the force between two polarizable atoms [3]. Although van der Waals forces are only relevant at small (micron scale) distances, they have been extensively measured (see, e.g., Refs. [4,5]). With recent advances in measurement techniques, including the microelectromechanical systems (MEMS) framework [6], Casimir-Polder forces become accessible in many other interesting conditions.

Due to the dominance of van der Waals forces in nanoscale devices, there has been much interest in controlling such forces. The full Lifshitz theory for van der Waals forces [2] shows their dependence on the electrical properties of the materials involved. Consequently, the possibility of tuning a material’s electric properties opens up the possibility of tuning fluctuation-induced interactions. This principle has been demonstrated in a number of experimental setups, for instance, by changing the charge carrier density of materials via laser heating, which of course engenders a consequent change in electrical properties [8]. There is also experimental evidence of the reduction of van der Waals forces for refractive-index-matched colloids [9–11]. The question of forces in external fields, electric and magnetic, has been studied in several articles [12–19]. When applying external fields, materials with a nonlinear electric response (which exhibit “nonlinear optics”) open up a variety of possibilities; these possibilities are absent in purely linear systems where the external field and fluctuating field are merely superimposed. Practically, metamaterials are promising candidates for Casimir force modulation, as they can exhibit strongly nonlinear optical properties [20,21] and their properties can be tuned by external fields [22]. The nature and description of fluctuation-induced effects in nonlinear systems are still under active research [23–26], including critical systems, where the underlying phenomenon is per se nonlinear [11]. For example, in Ref. [26], it was shown that nonlinear properties may alter Casimir forces over distances in the nanoscale. However, in the presence of only a small number of explicit examples, more research is needed to understand the possibilities opened up by nonlinear materials.

In this article, we consider an analytically solvable model for (anharmonic) point particles with strongly nonlinear responses. This is achieved by introducing a maximal, limiting value for the polarization of the particles, i.e., by confining the polarization vector in anharmonic potential wells. Casimir forces in such systems appear to be largely unexplored, even at the level of two-particle interactions. We find that strong external electric fields can be used to completely suppress the Casimir force in such systems. We discuss the stark difference of forces compared with the case of harmonic dipoles and give an asymptotic formula for the force in strong external fields, which we believe is valid in general if the involved particles have a maximal value for the polarization (saturate). In order to allow for analytical results, we restrict our analysis to the classical (high temperature) limit. However, similar effects are to be expected in quantum (low temperature) cases.

We start by computing the Casimir force for harmonic dipoles in an external field in Sec. II, where in Sec. II B we discuss the role of the angle between the field and the center-to-center axis. In Sec. III A we introduce the nonlinear (anharmonic) well model and compute the Casimir force in an external field in Sec. III C. We finally give an asymptotic expression for high fields in Sec. III D.

II. FORCE BETWEEN HARMONIC DIPOLES IN A STATIC EXTERNAL FIELD

A. Model

Classical van der Waals forces can be described by use of quadratic Hamiltonians describing the polarization of the
particles involved [27–29]. We introduce the system comprising two dipole carrying particles having the Hamiltonian,

\[ H^{(h)} = H_1^{(h)} + H_2^{(h)} + H_{\text{int}}, \]  

(1)

\[ H_i^{(h)} = \frac{p_i^2}{2\alpha} - p_i \cdot E, \]  

(2)

\[ H_{\text{int}} = -2k[3(p_i \cdot \hat{R})(p_j \cdot \hat{R}) - p_i \cdot p_j], \]  

(3)

where \( p_i \) is the instantaneous dipole moments of particle \( i \). Here \( \alpha \) denotes the polarizability, where, for simplicity of presentation, we choose identical particles. The external, homogeneous static electric field \( E \) couples to \( p_i \) in the standard manner. The term \( H_{\text{int}} \) describes the nonretarded dipole-dipole interaction in \( d = 3 \) dimensions with the coupling constant

\[ k = \frac{1}{4\pi \varepsilon_0} R^{-3}, \]  

(4)

where \( R = |R| \) with \( R \) the vector connecting the centers of the two dipoles, while \( \hat{R} \) denotes the corresponding unit vector. Since we are considering purely classical forces, retardation is irrelevant. Here \( \varepsilon_0 \) is the vacuum permittivity, and we use SI units. Inertial terms are irrelevant as well and have been omitted. (Since the interaction does not depend on, e.g., the change of \( p_i \) with time, inertial parts can be integrated out from the start in the classical setting.)

B. Casimir force as a function of the external field

The force \( F \) for the system given in Eqs. (1)–(3), at fixed separation \( R \), can be calculated from (as the external electric field is stationary, the system is throughout in equilibrium)

\[ F = \frac{1}{\beta} \partial_R \ln Z, \]  

(5)

where \( Z = \int d^3p_i \int d^3p_j \exp(-\beta H) \) is the partition function, with the inverse temperature \( \beta = 1/k_B T \) and \( H \) is the Hamiltonian of the system. By using the coupling constant \( k \) from Eq. (4), this may also be written as

\[ F = \frac{1}{\beta} \partial_{kR} \frac{\partial_k Z}{Z}. \]  

(6)

Furthermore, we are interested in the large separation limit, and write the standard series in inverse center-to-center distance (introducing \( R = |R| \)),

\[ F = \frac{1}{\beta} \partial_{kR} \left( \frac{\partial_k Z}{Z} \right)_{k=0} + \frac{1}{\beta} \partial_{kR} k \left[ \left( \frac{\partial_k^2 Z}{Z} \right) - \left( \frac{\partial_k Z}{Z} \right)^2 \right]_{k=0} + O(R^{-10}). \]  

(7)

In this series, the first term is of order \( R^{-4} \), while the second is of order \( R^{-7} \). The external electric field induces finite (average) dipole moments. For an isolated particle, this is (index \( 0 \) denoting an isolated particle, or \( k = 0 \))

\[ \langle p_i \rangle_0 = \frac{1}{k} \int d^3p_i \exp(-\beta H) p_i. \]  

(8)

\[ \langle p_i \rangle_0 = \frac{1}{k} \int d^3p_i \exp(-\beta H) p_i. \]  

(9)

For the case of harmonic particles, Eq. (2), this naturally gives

\[ \langle p_i \rangle_0 = \alpha E. \]  

(10)

The mean dipole moments of the isolated particles in Eq. (9), induced by the external electric field, give rise to a force decaying as \( R^{-4} \), i.e., the first term in Eq. (7). This can be made more explicit by writing

\[ \left( \frac{\partial_k Z}{Z} \right)_{k=0} = 2 \langle p_i \rangle_0 \cdot \langle p_j \rangle_0 - 6 \langle (p_i) \cdot \hat{R} \rangle \langle (p_j) \cdot \hat{R} \rangle. \]  

(10)

Representing a force decaying as \( R^{-4} \), this term dominates at large separations. From Eq. (10), the dependence on the angle between \( E \) and \( R \) becomes apparent. The induced force to order \( R^{-4} \) can be either attractive (e.g., \( R \parallel E \)) or repulsive (e.g., \( R \perp E \)) [30]. We are aiming at reducing the Casimir force through the electric field, and thus, term by term, try to obtain small prefactors. The considered term \( \sim R^{-4} \) is readily reduced by choosing \( \hat{R} \cdot \hat{E} = -\frac{1}{\sqrt{3}} \), for which this term is exactly zero, \( \left( \frac{\partial_k Z}{Z} \right)_{k=0} = 0 \). See the inset of Fig. 1 for an illustration. In the following sections we will thus study the behavior of the term \( \sim R^{-7} \) as a function of the external field, keeping this angle throughout.

C. Force for the angle \( \hat{R} \cdot \hat{E} = \frac{1}{\sqrt{3}} \)

For \( \hat{R} \cdot \hat{E} = \frac{1}{\sqrt{3}} \), the force is of order \( R^{-7} \) for large \( R \), and reads

\[ F|_{\hat{R} \cdot \hat{E} = \frac{1}{\sqrt{3}}} = \frac{\partial_{kR}^2}{2\beta} \left( \frac{\partial_k^2 Z}{Z} \right)_{k=0} + O(R^{-10}). \]  

(11)

The discussion up to here, including Eq. (11), is valid generally, i.e., for any model describing individual symmetric particles, where the induced polarization is in the direction of the applied field. For the case of harmonic dipoles, i.e., for Eq. (2), we denote \( F = F_0 \). Calculating \( \left( \frac{\partial_k Z}{Z} \right)_{k=0} \) for this case yields a result which is partly familiar from the case of harmonic dipoles in the absence of external fields.

FIG. 1. Casimir force between harmonic dipoles as a function of the strength of the external field. The angle between the field and the center-to-center vector \( R \) is chosen \( \varphi = \arccos(\frac{1}{\sqrt{3}}) \). The force component decaying with \( \sim R^{-4} \) [discussed after Eq. (10)] then vanishes, so that the force decays as \( \sim R^{-7} \).
Also (34) below. The term proportional to the number of parameters to a minimum, we additionally take $E$ to be

$$E = \frac{\alpha^2}{(4\pi\varepsilon_0)^2} R^{-7}. \quad (13)$$

Again, for zero field, $E \to 0$, this is in agreement with the Casimir-Polder force in the classical limit [27], given by $F_0$.

As the field is applied, the force increases, being proportional to $E^2$ for $\alpha \beta E^2 \gg 1$. This is due to interactions of a dipole induced by the $E$ field with a fluctuating dipole [compare also (34) below]. The term proportional to $E^2$ is naturally independent of $T$. The force as a function of external field is shown in Fig. 1.

The Casimir force given by Eq. (12) is thus tunable through the external field, but it can only be increased due to the square power law. While this might be useful for certain applications, we shall in the following investigate the case of highly nonlinear particles. The fact that the force in Eq. (13) is proportional to $\alpha^2$ suggests that reduction of the force could be achieved, if the polarizabilities were dependent on the external field. In the next section, we will investigate a model for saturating particle dipole moments, where indeed the forces can be suppressed.

III. FORCE BETWEEN SATURATING DIPOLES IN AN EXTERNAL FIELD

A. Model: Infinite wells

The response of a harmonic dipole to an external field is by construction linear for any value of the field [see Eq. (9)], and the polarization can be increased without bound. We aim here to include saturation by introducing a limit $P$ for the polarization, such that $|p_i| < P$ at all times and for all external fields. This can be achieved by modifying the Hamiltonian in Eq. (2), assigning an infinite value for $|p_i| > P$. The potential for $|p_i|$ obtained in such a way is illustrated in Fig. 2.

As we aim to study the effect of saturation, while keeping the number of parameters to a minimum, we additionally take the limit $\alpha \to \infty$. This yields an infinite well potential (see the lower curves of Fig. 2 for the approach of this limit),

$$H_i^{(w)} = \begin{cases} [−p_i \cdot E, |p_i| < P, \\ \infty, \end{cases} \quad \text{otherwise.} \quad (14)$$

Such models have been studied extensively in different contexts, as, e.g., asymmetric quantum wells of various shapes [31–33], two-level systems with permanent dipole moments [34], and dipolar fluids [35]. These systems are also known to be tunable with an external electric field [36,37]. However, the Casimir effect has not been investigated.

This model, for example, mimics free electrons confined to a spherical volume, such as in a perfectly conducting, neutral sphere. The maximum value for the dipole moment in this case is the product of the radius and the total free charge of the sphere. The charge distribution in a sphere has, additionally to the dipole moment, higher multipole moments, e.g., quadrupolar. For a homogeneous external field, the Hamiltonian in Eq. (14) is, however, precise, as higher multipoles couple to spatial derivatives (gradients) of the field [30], and only the dipole moment couples to a homogeneous field. Also, the interaction part, Eq. (3), contains, in principle, terms with higher multipoles. These do not, however, play a role for the force at the order $R^{-7}$.

B. Polarization and polarizability

We start by investigating the polarization of an individual particle as a function of the field $E$, resulting from Eq. (14), which is defined in Eq. (8). It can be found analytically,

$$\langle p_i \rangle_0 = Q(\beta EP) P \hat{E}, \quad (15)$$

$$Q(x) = \frac{1}{x} \frac{(x^2 - 3x + 3)e^{x^2} - x^2 - 3x - 3}{(x-1)e^{x^2} + x + 1}. \quad (16)$$

Note that the product $\beta EP$ is dimensionless. For a small external field, we find the average polarization is given by

$$\langle p_i \rangle_0 = \frac{1}{2} \beta P^2 E + O(E^3). \quad (17)$$

We hence observe, as expected, that for a small field the particles respond linearly, with a polarizability $\alpha_0 \equiv \frac{1}{2} \beta P^2$. This polarizability depends on temperature, as it measures how strongly the particles’ thermal fluctuations in the well are perturbed by the field. We may now give another interpretation of the limit $\alpha \to 0$ in Fig. 2. In order to behave as a “perfect” well, the curvature, given by $\alpha^{-1}$, must be small enough to fulfill $\alpha \gg \alpha_0$.

The normalized polarization [i.e., $Q(\beta EP) = \frac{\langle |p_i| \rangle}{\langle p_i \rangle_0}$] is shown in Fig. 3 as a function of external field. For small values of $E$, one sees the linear increase, according to Eq. (17). In the large field limit, the polarization indeed saturates to $P \hat{E}$. The dimensionless axis yields the relevant scale for $E$, which is given through $(\beta P)^{-1}$. At low temperature (or large $P$), saturation is approached already for low fields, while at high temperature (or low $P$), large fields are necessary for saturation.

Another important quantity related to the polarization is the polarizability, which is a measure of how easy it is to induce or change a dipole moment in a system. For harmonic
The amplitudes for parallel and perpendicular polarizability grow linearly, the polarizability is independent of $E$. In general, we can write

$$\alpha_{ij} = \frac{\partial\langle p_i \rangle}{\partial E_j}. \quad (18)$$

Note that this derivative is not necessarily taken at zero field $E$, so that $\alpha_{ij}$ is a function of $E$. Indices $i$ and $j$ denote the components of vectors (in contrast to previous notation). The polarizability tensor as defined in Eq. (18) is measured in the absence of any other particle (in other words, at coupling $k = 0$). $\alpha_{ij}$ can be deduced directly from the function $Q$ in Eq. (16). In general, we can write

$$\alpha_{ij}(E, P) = A_{ij}(\beta EP)\alpha_0. \quad (19)$$

Recall the zero-field polarizability is given as $\alpha_0 \equiv \frac{1}{2}\beta P^2$ [see Eq. (17)]. For the isolated particle, the only special direction is provided by the external field $E$, and it is instructive to examine the polarizability parallel and perpendicular to it. Taking, for example, $E$ along the $z$ axis, the corresponding dimensionless amplitudes $A_1 = A_{zz}$ and $A_\perp = A_{xx} = A_{yy}$ are

$$A_1(x) = \frac{5}{3} \frac{d}{dx} Q(x), \quad (20)$$

$$A_\perp(x) = 5 \frac{1}{x} Q(x). \quad (21)$$

The amplitudes for parallel and perpendicular polarizability are also shown in Fig. 3. The direct connection with the polarization is evident. For small fields, where the polarization grows linearly, the polarizability is independent of $E$. Analytically,

$$A_1(x) = 1 - \frac{3}{35} x^2 + O(x^3), \quad (22)$$

$$A_\perp(x) = 1 - \frac{1}{35} x^2 + O(x^3). \quad (23)$$

For large fields, i.e., when $\beta EP$ is large compared to unity, the polarizability reduces due to saturation effects. Asymptotically for large fields, the polarizability amplitudes are given as

$$A_1(x) = 10x^{-2} + O(x^{-3}), \quad (24)$$

$$A_\perp(x) = 5x^{-1} - 10x^{-2} + O(x^{-3}). \quad (25)$$

The parallel polarizability $\alpha_{||}$ falls off as $E^{-2}$ and the parallel polarizability $\alpha_{\perp}$ as $E^{-1}$. The different power laws may be expected, as near saturation, changing the dipole’s direction is a softer mode compared to changing the dipole’s absolute value.

C. Casimir force

The Casimir force between particles described by the well potential, Eq. (14), is computed from the following Hamiltonian,

$$H^{(w)} = H_1^{(w)} + H_2^{(w)} + H_{int}. \quad (26)$$

$$H_1^{(w)} = -\mathbf{p}_i \cdot \mathbf{E}, \quad |\mathbf{p}_i| < P,$$

$$\infty, \quad \text{otherwise.} \quad (27)$$

with the interaction potential $H_{int}$ given in Eq. (3). The discussion in Sec. II regarding the angle of the external field holds similarly here, i.e., Eq. (11) is valid and the force decaying as $R^{-4}$ vanishes for the angle $\hat{R} \cdot \hat{E} = \frac{1}{\sqrt{2}}$. Therefore, we continue by studying the $R^{-7}$ term at this angle. Using Eq. (11), the Casimir force can be found analytically,

$$F_w = f_w(\beta EP)F_0 + O(R^{-10}), \quad (28)$$

with the zero-field force

$$F_0 = -\frac{72}{\beta} \left(\frac{\alpha_0}{4\pi \varepsilon_0}\right)^2 R^{-7}. \quad (29)$$

and the dimensionless amplitude

$$f_w(x) = \frac{25}{3} \frac{1}{x^4} \left(\frac{1}{2} + 3 \sinh(x) - 3x \cosh(x)\right) \times \frac{\sinh(x)}{[x \cosh(x) - \sinh(x)]^2} \times \frac{x}{(2x^2 + 21)x \cosh(x) - 9(x^2 + 21) \sinh(x)]}. \quad (30)$$

Again, $\alpha_0 \equiv \frac{1}{2}\beta P^2$ is the zero-field polarizability [see Eq. (17)]. The force is most naturally expressed in terms of $F_0$, which is the force at zero field, equivalent to Eq. (13). The amplitude $f_w$ is then dimensionless and depends, as the polarization, on the dimensionless combination $\beta EP$.

The force is shown in Fig. 4. For zero external fields, the curve starts at unity by construction, where the force is given by $F_0$. The force initially increases for small values of $\beta EP$, in accordance with our earlier analysis of harmonic dipoles. After this initial regime of linear response, the Casimir force decreases for $\beta EP \gtrsim 1$, and, for $\beta EP \gg 1$, asymptotically approaches zero as $E^{-1}$,

$$F_w = -\frac{48P^3}{(4\pi \varepsilon_0)^2} R^{-7} E^{-1} + O(E^{-2}). \quad (31)$$

This behavior yields an enormous potential for applications: By changing the external field, the force can be switched on or off.
The asymptotic law in Eq. (31) gives another intriguing insight: For large fields, the force is independent of temperature. This is in contrast to the fact that (classical) fluctuation-induced forces in general do depend on temperature. This peculiar observation is a consequence of cancellations between factors of $\beta$, and might yield further possibilities for applications. This is demonstrated in Fig. 5, where we introduced a reference temperature $T_0$. Indeed, we see that for small values of $E$, the force does depend on temperature, while for large fields, the curves for different values of temperature fall on top of each other. As a remark, we note that $F_0$ is inversely proportional to temperature, in contrast to $F_0$ for harmonic particles in Eq. (13). This is because the zero-field polarizability depends on temperature for the well potentials considered here.

Regarding experimental relevance, it is interesting to note that, in a somewhat counterintuitive way, larger values of $P$ lead to stronger dependence on the external field $E$ (the important parameter is $\beta EP$). We thus expect that larger particles are better candidates for observing the effects discussed here. For example, for a gold sphere of radius 100 nm, we estimate $P = 5 \times 10^{-19}$ Cm, so that $\beta EP \sim 1$ for $E = 10$ mV/m at room temperature.

### D. Asymptotic formula for high fields

What is the physical reason for the decay of the force for large field $E$ observed in Fig. 4? For large values of $\beta EP$, the force may be seen as an interaction between a stationary dipole and a fluctuating one. This is corroborated by a direct computation of the force between a stationary dipole $\mathbf{q}$, pointing in the direction of the electric field, and a particle with the Hamiltonian

$$H_{10} = \frac{p^2}{2\alpha_\perp} + \frac{p^2}{2\alpha_\parallel} - \mathbf{p} \cdot \mathbf{E},$$

where “perpendicular” and “parallel” refer to the direction of the $E$ field as before. The two such hypothetical particles interact via the Hamiltonian

$$H_{int}^{(0)} = -2k[3(\mathbf{p} \cdot \hat{\mathbf{R}})(\mathbf{q} \cdot \hat{\mathbf{R}}) - \mathbf{p} \cdot \mathbf{q}].$$

Choosing the angle between $\mathbf{R}$ and $\mathbf{E}$ as before, we find for the force between these particles (to leading order in $k$),

$$F_s = -24 \alpha_\perp q^2 \left( \frac{1}{4\pi \epsilon_0} \right)^2 R^{-7}.$$  \hspace{1cm} (34)

This result can be related to Eq. (31). Substituting $\mathbf{q} = \mathbf{P} \hat{\mathbf{E}}$, the value at saturation, and $\alpha_\perp = 5/(\beta EP)\alpha_0 = P/E$ [using the leading term for large field from Eq. (25)], we find

$$F_s = -24 \frac{\alpha_\perp}{E} \left( \frac{1}{4\pi \epsilon_0} \right)^2 R^{-7}.$$  \hspace{1cm} (35)

This is identical to Eq. (34), except for a factor of 2. This is expected, as this factor of 2 takes into account the force from the first fixed dipole interacting with the second fluctuating one and vice versa. We have thus demonstrated that Eq. (34) may be used to describe the behavior of the force for large values of $E$. The importance of this observation lies in the statement, that such reasoning might be applicable more generally: in the case of more complex behavior of $p(E)$, i.e., more complex (or realistic) particles. We believe that the value of $q$ at saturation and the polarizability $\alpha_\perp$ near saturation can be used to accurately predict the force in the limit of large external fields via Eq. (34).

### IV. SUMMARY

We have demonstrated how the classical Casimir-Polder force between two saturating dipoles can be suppressed by applying an external static electric field. Of special interest is the angle $\varphi = \arccos(\frac{1}{\sqrt{3}})$ between the external field and the vector connecting the dipoles, for which the deterministic dipole-dipole interaction vanishes. The remaining “Casimir-Polder” part can then be tuned and is arbitrarily suppressed at large values of external fields due to the vanishing polarizability. The force in this case decays as $E^{-1}$. This is in strong contrast to harmonic dipoles, which experience an increase of the force in the presence of an external field, growing with
We also provided a simple formula to estimate the force between particles under strong fields. It would be interesting to extend the results here to macroscopic objects composed of such dipole carrying particles, where multbody effects will potentially change the physics for dense systems. However, for dilute systems, where the pairwise approximation of van der Waals forces is accurate, the results obtained here are directly applicable and thus the modulation of Casimir or van der Waals forces predicted here will apply to a certain extent. Of course, an important main difference in more than two-body systems is that the deterministic component of the interaction cannot be obviously canceled by a uniform electric field, as there is more than one center-to-center vector, denoted by $R$ in this article, separating the interacting dipoles.

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