

It is predicted that in force microscopy the quantum fluctuations responsible for the Casimir force can be directly observed as temperature-independent force fluctuations having spectral density $9\pi/(40 \ln(4/e)) \hbar |\delta k|$, where \hbar is Planck's constant and δk is the observed change in spring constant as the microscope tip approaches a sample. For typical operating parameters the predicted force noise is of order 10^{-18} Newton in one Hertz of bandwidth. The Second Law is respected via the fluctuation-dissipation theorem. For small tip-sample separations the cantilever damping is predicted to increase as temperature is reduced, a behavior that is reminiscent of the Kondo effect.

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The Casimir force is the mean force between two objects that is generated by quantum fluctuations [1]. In a recent review article [2], Barton notes: "It is strange that for nearly half a century after Casimir no curiosity has been displayed regarding the fluctuations [of the Casimir force] about the mean." This lack of curiosity is understandable in view of the prevailing opinion, as summarized in [2], that Casimir fluctuations are "far too small to detect with any traditionally contemplated Casimir-type apparatus."

In this article we venture a contrary prediction—that Casimir force fluctuations are large enough to be directly detected by force microscopes, and that these fluctuations provide a fundamental limit to force microscope sensitivity which is stringent enough to be significant in practical applications [11]. We obtain this prediction via a strategy advocated by Kupiszewska [3]:

The standard macroscopic quantum theory for nonhomogenous media . . . refers to a medium described by a constant refractive index. Although useful for many applications, this approach, as well as all other approaches neglecting losses, is generally incorrect. It is well known that the dielectric function must satisfy the Kramers-Kronig relations, otherwise causality would be violated. According to Kramers-Kronig relations, the imaginary part of a realistic, frequency-dependent dielectric function must not vanish, and that implies the dissipation of radiation energy. Therefore, a complete theory will have to include not only the field and the atoms, but also a system that absorbs energy, usually called a heat bath or reservoir.

In implementing Kupiszewska's program, we will consider the two force microscope geometries shown in Fig. 1. Both geometries assume a spherical cantilever tip. We confine our attention to experiments conducted *in vacuo* at cryogenic temperatures [11], because such experiments



FIG. 1. Two force microscope geometries: at left, tip vibration normal to the sample plane; at right, tip vibration in the sample plane.

have the force sensitivity required to directly observe the predicted Casimir fluctuations.

Our discussion centers upon four parameters which change when the cantilever tip approaches the sample—our goal is to predict these changes. The four parameters are: (a) the resonant frequency ω_0 , (b) the spring constant k , (c) the cantilever resonant quality Q , and (d) the force noise spectral density S_f .

We begin by describing how these parameters are measured. In typical experiments [11], the tip is brought near the sample and the Brownian motion $x(t)$ of the cantilever tip is observed interferometrically. In well-designed experiments $x(t)$ is dominated by thermal noise, such that the cantilever motion is in thermal equilibrium with $k\langle x^2 \rangle = k_B T$, where $\langle x^2 \rangle$ is the mean square tip displacement, k_B is Boltzmann's constant, and T is the ambient temperature. In such experiments the autocorrelation of $x(t)$ is exponential:

$$\langle x(t)x(t+\tau) \rangle_t = \langle x^2 \rangle e^{-\omega_0\tau/(2Q)} \cos(\omega_0\tau), \quad (1)$$

where $\langle \rangle_t$ denotes a time average. The autocorrelation

thus determines the parameters $\langle x^2 \rangle$, ω_0 , and Q . From them, it is routine practice to: (a) infer the spring constant via $k = m\omega_0^2$, with m the motional mass of the cantilever, (b) verify that thermodynamic equilibrium is respected by checking that $k\langle x^2 \rangle = k_B T$, and (c) calculate a Langevin force spectral density S_f via

$$\begin{aligned} S_f &\equiv \int_{-\infty}^{\infty} d\tau e^{-i\omega\tau} \langle f(t)f(t+\tau) \rangle \\ &= \frac{2k^2}{Q\omega_0} \langle x^2 \rangle = \left(\frac{m\omega_0}{Q} \right) 2k_B T. \end{aligned} \quad (2)$$

The term $m\omega_0/Q$ in (2) is recognizably the damping coefficient of a mechanical oscillator; (2) thus represents the fluctuation-dissipation theorem [4] as it applies to force microscope experiments. By this method, or inconsequential variants thereof, the four parameters ω_0 , Q , k , and S_f are routinely measured in force microscopy.

For tip vibration normal to the sample plane (as in Fig. 1 at left), the spring constant k decreases as the tip approaches the sample. Our main prediction, which is derived in the second half of this article, is that the observed change in spring constant δk will be accompanied by an increase in force noise δS_f according to

$$\delta S_f = \frac{9\pi}{40 \ln(4/e)} \hbar (-\delta k). \quad (3)$$

Then from (2) and (3), the dynamical cantilever damping $m\omega_0/Q$ is predicted to increase according to

$$\delta \left(\frac{m\omega_0}{Q} \right) = \frac{1}{2k_b T} \delta S_f = \frac{9\pi}{80 \ln(4/e)} \frac{\hbar}{k_B T} (-\delta k), \quad (4)$$

thus ensuring that the Casimir fluctuations respect thermodynamic equilibrium, such that $k\langle x^2 \rangle = k_B T$ for all values of δk . Note that these predictions involve only experimentally measured quantities and Planck's constant—there are no model-dependent parameters.

Are the predicted force fluctuations large enough to observe directly? From (3), we compute that a spring constant shift $\delta k = -2.6 \times 10^{-3} \text{ N/m}$ will be associated with a temperature-independent Casimir force noise of $10^{-18} \text{ N}/\sqrt{\text{Hz}}$. Spring constant shifts of this magnitude are commonly observed in force microscopy. Recent experiments have demonstrated force noise levels of order $10^{-17} \text{ N}/\sqrt{\text{Hz}}$, and if force sensitivity continues to improve [11] then it is reasonable to expect that the predicted Casimir fluctuations will become a dominant noise mechanism in future force microscope experiments.

To describe experiments in which tip vibration is in the plane of the sample—as in Fig. 1 at right—we introduce a length scale l defined such that an attractive Casimir force f exerted between tip and sample generates a change in spring constant $\delta k = f/l$. To calculate l explicitly, let $\phi(z)$ be the modal eigenfunction of the cantilever, with z the coordinate along the cantilever length L , normalized such that $\phi(L) = 1$. Then l is given by

$$l^{-1} = \int_0^L dx \left[\frac{\partial \phi(x)}{\partial x} \right]^2. \quad (5)$$

Typically l/L is of order unity. Letting h be the tip-to-sample separation distance, the predicted increase in force noise as the tip approaches the sample is

$$\delta S_f = \frac{3\pi}{160 \ln(4/e)} \frac{l}{h} \hbar \delta k. \quad (6)$$

There is no minus sign in this equation, in contrast to (3), because the end-on geometry shown at right in Fig. 1 yields a positive δk at close tip-sample separation. Then (2) and (6) yield the predicted damping increase:

$$\delta \left(\frac{m\omega_0}{Q} \right) = \frac{1}{2k_b T} \delta S_f = \frac{3\pi}{320 \ln(4/e)} \frac{\hbar}{k_B T} \frac{l}{h} \delta k. \quad (7)$$

Assuming δk is independent of temperature to leading order—which is a reasonable assumption for Casimir forces—the predicted cantilever damping varies *inversely* with temperature, according to (4) and (7). Such inverse relations are uncommon in physics but they are not unknown; the Kondo effect is an example.

It remains only to derive (3) and (6) by the program of Kupiszewska. We will not hesitate to make brutal simplifying approximations along the way, with a view toward obtaining results in closed form. Furthermore, we will finesse various model-dependent parameters by showing that they appear in δS_f and δk in such a manner that the ratio $\delta S_f/(\delta k)$ is parameter-independent. By this strategy we can reasonably hope to obtain final results which have broader validity than the underlying model from which they derive.

Following Kupiszewska [3], we model individual atoms as independent harmonic oscillators. By an appropriate scaling of variables the interaction of a tip atom a with a sample atom b can be described by the Hamiltonian

$$\begin{aligned} H &= \frac{1}{2} \omega_a (p_a^2 + q_a^2) + \frac{1}{2} \omega_b (p_b^2 + q_b^2) \\ &\quad - \beta_{ab} q_a q_b + (\text{heat bath}). \end{aligned} \quad (8)$$

Here ω_a and ω_b are atomic oscillator frequencies and β_{ab} is a dipole coupling whose strength and spatial dependence are discussed later. The operators $\{q_a, p_a, q_b, p_b\}$ obey the usual commutation relations $[q_i, p_j] = i\hbar \delta_{ij}$, and it will turn out that no other information need be specified about their physical nature. We specify the heat bath as the unique independent oscillator (IO) model of Ford *et al.* [5] that induces linear damping in the Heisenberg picture equations of motion:

$$\ddot{q}_a + \Gamma_a \omega_a \dot{q}_a + \omega_a^2 q_a = F_a(t) + \beta_{ab} q_b \quad (9)$$

$$\ddot{q}_b + \Gamma_b \omega_b \dot{q}_b + \omega_b^2 q_b = F_b(t) + \beta_{ab} q_a. \quad (10)$$

Here $F_a(t)$ and $F_b(t)$ are operator-valued Langevin forces which originate in the heat bath, and $\{\Gamma_a, \Gamma_b\}$ are damping rates. We pause to note that our conventions for correlation and power spectral density are those of Balescu's textbook [4]:

$$C_{AB}(\tau) \equiv \frac{1}{2} \langle A(t)B(t+\tau) + B(t+\tau)A(t) \rangle$$

$$S_A(\omega) \equiv \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} C_{AA}(\tau),$$

with $\langle \rangle$ an expectation over heat bath variables. Then as shown by Ford *et al.* [5], the assumption of linear damping in (9-10) uniquely determines the Langevin force autocorrelation to be

$$C_{F_i F_j}(\tau) = \delta_{ij} \frac{\Gamma_i \omega_i}{2\pi} \int_{-\infty}^{\infty} d\omega \hbar \omega \coth\left(\frac{\hbar \omega}{2k_B T}\right) e^{-i\omega\tau}. \quad (11)$$

The resulting model of atomic fluctuations resembles Kupiszewska's model of gauge field fluctuations quite closely. Kupiszewska's model integrates over matter fields to obtain equations in which only gauge fields appear. Our strategy is the opposite: we have integrated over the longitudinal gauge fields which generate the atomic dipole coupling [6], such that (9–11) contain only matter fields. The two approaches are formally equivalent, because in the real world gauge fluctuations and matter fluctuations are inseparable, such that either can be regarded as the fundamental dynamical variable.

We now wish to compute the pairwise Casimir force \mathbf{f}_{ab} between two atoms, the associated spring constant \mathbf{k}_{ab} , and the force spectral density $\mathbf{S}_{\mathbf{f}_{ab}}(\omega)$. The force is given in terms of the gradient of the dipole coupling by [4]

$$(\mathbf{f}_{ab})_i = (\nabla_i \beta_{ab}) \langle q_a q_b \rangle, \quad (12)$$

from which \mathbf{k}_{ab} and $\mathbf{S}_{\mathbf{f}_{ab}}(\omega)$ follow immediately as

$$(\mathbf{k}_{ab})_{ij} = -\nabla_i (\mathbf{f}_{ab})_j \quad (13)$$

$$(\mathbf{S}_{\mathbf{f}_{ab}}(\omega))_{ij} = (\nabla_i \beta_{ab}) (\nabla_j \beta_{ab}) S_{(q_a q_b)}(\omega), \quad (14)$$

where the spectral density $S_{(q_a q_b)}(\omega)$ is given by

$$S_{(q_a q_b)}(\omega) = \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} C_{(q_a q_b)(q_a q_b)}(\tau). \quad (15)$$

The only dynamical quantities that appear in (12–15) are the expectation $\langle q_a q_b \rangle$ and the autocorrelation $C_{(q_a q_b)(q_a q_b)}(\tau)$. Our next step, therefore, is to compute these quantities to leading and next-to-leading order in $\{\beta_{ab}, \Gamma_a, \Gamma_b\}$, which are assumed small compared to $\{\omega_a, \omega_b\}$. Physically, the atomic oscillators are assumed to be weakly coupled by the Casimir interaction and underdamped by the ambient heat bath.

As described in [5] and [7], any desired correlation involving $q_a(t)$ and $q_b(t)$ can be explicitly computed by solving (9–11) in the Fourier domain. For $\langle q_a q_b \rangle$ an uncomplicated Fourier integration yields

$$\langle q_a q_b \rangle \stackrel{\tau \rightarrow 0}{=} \frac{\hbar \beta_{ab}}{2(\omega_a + \omega_b)} + \left[\frac{\hbar \beta_{ab} \omega_a \omega_b}{2\pi(\omega_a^2 - \omega_b^2)^2} \times \left(\Gamma_a \left(1 - \frac{\omega_a^2}{\omega_b^2} + \ln\left(\frac{\omega_a^2}{\omega_b^2}\right)\right) + (a \leftrightarrow b) \right) \right], \quad (16)$$

plus $\mathcal{O}(\beta^3, \beta \Gamma^2)$ terms. The zero of the denominator in (16) for $\omega_a = \omega_b$ cancels against a zero of the numerator, such that $\langle q_a q_b \rangle$ is finite for all nonzero ω_a and ω_b . Setting $\Gamma_a = \Gamma_b = 0$ in (16), we recover the same expression for $\langle q_a q_b \rangle$ as is obtained from the ground state of the Hamiltonian with the heat bath turned off.

These results establish that heat bath damping does not alter the interatomic Casimir force in leading order, as is physically reasonable.

With regard to the autocorrelation $C_{(q_a q_b)(q_a q_b)}(\tau)$ in (15), the Gaussian property of the Langevin forces, as discussed by Ford *et al.* [5], allows it to be written as the product of two simpler autocorrelations:

$$C_{(q_a q_b)(q_a q_b)}(\tau) = C_{q_a q_a}(\tau) C_{q_b q_b}(\tau) + \mathcal{O}(\beta_{ab}^2). \quad (17)$$

In turn, $C_{q_a q_a}(\tau)$ is calculated by solving (9–11) in the Fourier domain in a manner that precisely parallels the calculations of Li *et al.* [7]. The result is:

$$C_{q_a q_a}(\tau) \stackrel{\tau \rightarrow 0}{=} \frac{\hbar}{2} \int_a^\infty d\omega \frac{\Gamma_a \omega_a \omega \cos(\omega\tau)}{|\omega_a^2 - \omega^2 + i\Gamma_a \omega|^2} + \mathcal{O}(\beta_{ab}^2) \quad (18)$$

$$= \frac{\hbar}{2} \frac{\omega_a}{\bar{\omega}_a} \left[e^{-\Gamma_a |\tau|/2} \cos(\bar{\omega}_a \tau) + \frac{2}{\pi} \text{Im} g\left(\left(\bar{\omega}_a + \frac{i\Gamma_a}{2}\right)|\tau|\right) \right] + \mathcal{O}(\beta_{ab}^2) \quad (19)$$

$$= \frac{\hbar}{2} e^{-\Gamma_a |\tau|/2} \cos(\bar{\omega}_a \tau) + \mathcal{O}(\Gamma_a, \beta_{ab}^2). \quad (20)$$

By substituting $a \rightarrow b$ we obtain $C_{q_b q_b}(\tau)$. Here $\bar{\omega}_a \equiv (\omega_a^2 - \Gamma_a^2/4)^{1/2}$ is assumed real and positive, and $g(z)$ is the exponential integral [8] defined by

$$g(z) \equiv \int_0^\infty dt \frac{\cos(t)}{t+z}. \quad (21)$$

The term $\text{Im} g\left(\left(\bar{\omega}_a + i\Gamma_a/2\right)|\tau|\right)$ in (19) can be proved to be monotonic in $|\tau|$, with initial value $-\tan^{-1}(\Gamma_a/(2\bar{\omega}_a))$ and asymptotic value $-\Gamma_a \bar{\omega}_a / (\tau^2 \omega_a^4)$. This term therefore describes squeezing of the quantum zero-point motion by the heat bath damping—a phenomenon which is physically to be expected.

The engineering import of (20) is that even at zero temperature, where classical oscillators exhibit zero noise, the zero-point motion of a lightly damped ($\Gamma_a \ll \omega_a$) quantum oscillator carries $\hbar/2$ of noise power within a bandwidth Γ_a centered on a carrier frequency ω_a .

Since each atom in our model is coupled to an independent heat bath, and is dynamically independent of adjacent atoms, we can obtain the total Casimir force and force noise by summing the atomic interactions pairwise. For definiteness, we assume a Debye distribution of atomic frequencies $\{\omega_a, \omega_b\}$, such that $p(\omega) = 3\omega^2/\omega_D^3$, where ω_D is a Debye frequency—any other broad-band frequency distribution would yield similar results. Then we frequency-average $\langle q_a q_b \rangle$ from (16) and $S_{(q_a q_b)}(\omega)$

from (15), (17), and (20) to obtain to leading order in $\{\beta_{ab}, \Gamma_a, \Gamma_b\}$:

$$\begin{aligned} \langle q_a q_b \rangle^{av} &= \int_0^{\omega_D} d\omega_a d\omega_b p(\omega_a) p(\omega_b) \langle q_a q_b \rangle \\ &= \frac{9 \ln(4/e)}{10} \frac{\hbar \beta_{ab}}{\omega_D} \end{aligned} \quad (22)$$

$$\begin{aligned} S_{(q_a q_b)}^{av}(\omega) &= \int_{-\infty}^{\infty} d\tau \int_0^{\omega_D} d\omega_a d\omega_b \left[p(\omega_a) p(\omega_b) e^{i\omega\tau} \right. \\ &\quad \left. \times C_{(q_a q_b)(q_a q_b)}(\tau) \right] \stackrel{\omega \ll \omega_D}{\equiv} \frac{9\pi \hbar^2}{20\omega_D} \end{aligned} \quad (23)$$

In (23) we have assumed $\omega \ll \omega_D$, as appropriate for audio frequency force microscope experiments.

It remains only to integrate over the tip and sample volumes shown in Fig. 1. We specify that the tip and sample atoms have number density ρ_a and ρ_b respectively. The coupling β_{ab} is assumed to have a dipole dependence $\beta_{ab} = \kappa/|\mathbf{r}_{ab}|^3$, with \mathbf{r}_{ab} the atomic separation vector and κ the strength of the dipole interaction. Substituting (22–23) into (12–14) and integrating over the volume of a spherical tip of radius r that is separated by a gap h from a half-space sample, we obtain the total Casimir force, spring constant, and force noise as

$$\mathbf{f}^{tot} = -\hat{\mathbf{n}} \frac{\hbar \rho_a \rho_b \kappa^2}{\omega_D} \frac{3 \ln(4/e) \pi^2}{10} \frac{r^3}{h^2(2r+h)^2} \quad (24)$$

$$\begin{aligned} \mathbf{k}^{tot} &= -(\hat{\mathbf{n}} \otimes \hat{\mathbf{n}}) \frac{\hbar \rho_a \rho_b \kappa^2}{\omega_D} \\ &\quad \times \frac{6 \ln(4/e) \pi^2}{5} \frac{r^3(r+h)}{h^3(2r+h)^3} \end{aligned} \quad (25)$$

$$\begin{aligned} \mathbf{S}_{\mathbf{f}}^{tot} &= \left[\hat{\mathbf{n}} \otimes \hat{\mathbf{n}} + \frac{1}{24} (\mathbf{I} - \hat{\mathbf{n}} \otimes \hat{\mathbf{n}}) \right] \\ &\quad \times \frac{\hbar^2 \rho_a \rho_b \kappa^2}{\omega_D} \frac{27\pi^3}{100} \frac{r^3(r+h)}{h^3(2r+h)^3}. \end{aligned} \quad (26)$$

Here $(\mathbf{I})_{ij} \equiv \delta_{ij}$ is the identity matrix, $\hat{\mathbf{n}}$ is a unit vector normal to the sample surface, and $(\hat{\mathbf{n}} \otimes \hat{\mathbf{n}})_{ij} \equiv \hat{\mathbf{n}}_i \hat{\mathbf{n}}_j$ is an outer product. With the neglect of $\mathcal{O}(h/r)$ terms, as is reasonable for close tip-sample approach, our main results (3) and (6) follow immediately from (24–26).

Consistent with our stated goal of model independence, we need not specify numerical values for the parameters $\{\rho_a, \rho_b, \kappa, \omega_D, \Gamma_a, \Gamma_b, h, r\}$, because they do not appear in the final results. However, the dimensionless coefficients in (3) and (6) are weakly sensitive to the functional form of the assumed Debye distribution of atomic frequencies. Thus, for example, the coefficient $9\pi/(40 \ln(4/e)) \sim 1.83$ appearing in (3) is best regarded as a coefficient of order unity, whose precise value will depend on the material properties and shape of the tip and sample. These coefficients are well suited for experimental determination.

Arguably the two least realistic assumptions of our model are the assumed dynamical independence of adjacent atoms, and the coupling of each atom to an independent heat bath possessed of an infinite number of

degrees of freedom. The path to a more realistic model is clear but arduous. The heat bath model should be improved to describe realistic phonon and conduction band degrees of freedom, while taking into account the finite size of the tip, and electronic degrees of freedom in adjacent atoms should be realistically coupled. Both gauge fields and matter fields should be explicitly included in the Hamiltonian as in Kupiszewska's pioneering article [3]. The resulting field equations should be solved for realistic tip-sample geometries. Force fluctuations should be computed by the field-theoretic methods pioneered by Barton [9,10]. Ideally, the results should explicitly respect the fluctuation-dissipation theorem and should be expressed in a simple and physically transparent form.

Meeting these challenges will not be easy. Yet if the predictions of this article are experimentally confirmed, such that Casimir effects set the practical limits to force microscope sensitivity, then achieving a realistic understanding of these effects will become a matter of urgent practical consequence, in particular to the biomedical research community [11]. And if the predictions of this article are not confirmed, the question will be: why not?

In either case, it is certain that Casimir effects will continue to engage and delight the imaginations of the physicists and engineers in coming decades.

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