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## On the use of hydrogen switchable mirrors in Casimir force experiments

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**Abstract.** Hydrogen switchable mirrors (HSMs) are shiny metals that become optically transparent upon exposure to hydrogen. The Casimir force between HSMs is thus supposed to be stronger in air than in hydrogen. A few years ago, an experiment designed to measure this effect gave an unexpected result (Iannuzzi, Lisanti and Capasso 2004 *Proc. Natl Acad. Sci. USA* **101** 4019–23): no change of the force was observed upon hydrogenation. Qualitative arguments show that this result is reasonable if HSMs do not switch at long wavelengths, where no measurements of the dielectric function are available. Because the exact composition of the mirrors used in that experiment is not known, a more quantitative comparison of the data with theory is not possible. Still, calculations of the Casimir force in the presence of similar HSMs of known composition might provide new insights for the interpretation of the experimental result and may suggest precious hints for the development of future analogous experiments. In this paper, we present calculations of the Casimir attraction between Mg<sub>2</sub>Ni mirrors in air and in hydrogen. Our results clearly indicate that the modulation of the Casimir force with HSMs should be observable with modern set-ups.

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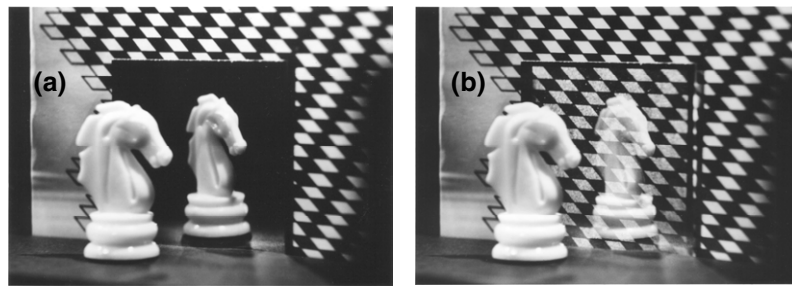
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**1. Introduction**

In the last decade, the literature dedicated to the Casimir effect [1] has been strongly influenced by a series of papers claiming the achievement of ultimate accuracy in the comparison of Casimir force experiments with theoretical predictions (see e.g. [2] and references therein). These papers have triggered a renewed interest in this field, stimulating a vivid discussion on the calculation of systematic errors of both experimental data and theoretical analysis. It is therefore not surprising that the large majority of modern experiments are performed using surfaces coated with thick, bulk-like metallic layers. From an experimental point of view, the use of metals offers the possibility to control and eliminate any residual electrostatic interaction that might disturb the measurements and increase the experimental uncertainty. Concerning the theoretical analysis of the data, the Casimir force between two objects is typically calculated by substituting the dielectric functions of their surfaces directly into the Lifshitz equation for van der Waals interaction [3]. The accuracy of the predicted results is thus strictly related to the degree of confidence with which those functions are known. The choice of bulk-like metallic films facilitates accurate calculations, because the dielectric functions of metals are known better than those of dielectrics. Measurements of Casimir forces between materials other than metals would thus not contribute to the discussion on the *accuracy issue*, giving rise to less impactful results.

It has been already recognized, however, that this trend represents a severe narrowing of the focus of modern literature [4]–[8]. The dependence of the Casimir force on the dielectric function of the interacting surfaces should not be considered only as an annoying detail that makes measurements and precise calculations more difficult. It should rather stimulate new experiments that explore the possibility of tailoring the Casimir effect using materials with suitably chosen dielectric functions.

A few years ago, in the attempt to emphasize the importance of studies that go beyond the accuracy issue, one of us (DI), in collaboration with M Lisanti and F Capasso, performed the first measurement of the Casimir force between hydrogen switchable mirrors (HSMs) [4]. HSMs are materials that alter their dielectric properties upon hydrogen absorption [9] (see figure 1). They change from reflective conductors to transparent semiconductors or insulators when they are transferred from air to hydrogen. The effect is reversible; when the mirror is put back in air, it switches again to the reflective state. Because of this change of their optical properties upon hydrogen absorption and desorption, HSMs offer in principle the possibility to observe a reversible, *in situ* change of the Casimir force between surfaces at fixed separation, a fascinating



**Figure 1.** An image of an HSM (a) before and (b) after exposure to hydrogen (courtesy of Ronald Griessen).

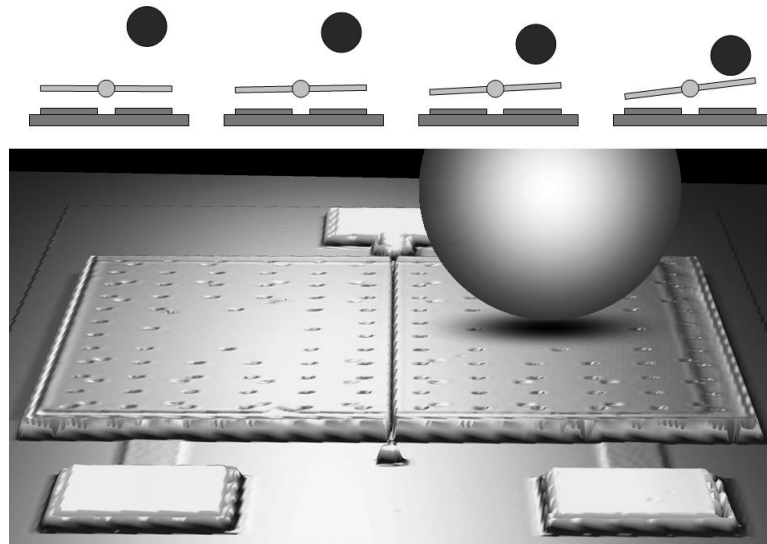
prospect from both a fundamental and technological point of view. According to the Lifshitz theory, the attraction between highly reflective metals is larger than the attraction between transparent dielectrics, which provide a less efficient confinement of the electromagnetic modes in the cavity formed by the two interacting surfaces. It should thus be possible to reduce the Casimir force between HSMs by exposing the mirrors to a hydrogen atmosphere; the force could then be brought back to higher values by replacing hydrogen with air. Surprisingly, the experiment reported in [4] was not capable of detecting any effect. In spite of the dramatic change of the optical properties of the mirrors, the Casimir force measured in hydrogen was not significantly different from that obtained in air. Due to the limited amount of information on the dielectric function of the HSMs used in the experiment, the authors could not present a rigorous theoretical calculation to compare with the data. Nevertheless, a mathematical exercise based on *ad hoc* materials with properly chosen dielectric functions made it possible to explain this unexpected result in terms of a counterintuitive property of the Lifshitz theory. Yet, a complete analysis of the magnitude of the effect is still missing, and one might wonder if there is a configuration for which a reversible change of the Casimir force between HSMs upon hydrogenation could be observed with modern experimental set-ups.

The goal of the present paper is to address this topic. First, we will briefly review the previous experiment and the mathematical arguments reported to explain the data. Then, we will discuss the switching properties of  $\text{Mg}_2\text{Ni}$  mirrors, HSMs with composition similar to the one used in the experiment. We will show that, although direct measurements of reflectivity and transmittivity are available only in a limited wavelength region, it is still possible to estimate the behaviour of the dielectric function over a much broader spectrum by means of a simultaneous analysis of Hall effect and resistivity data. Finally, we will present the results of Casimir force calculation obtained by substituting these dielectric functions in the Lifshitz equation. We will demonstrate that the decrease in the attraction after hydrogenation of the mirrors is expected to be smaller than the sensitivity of the experiment discussed above, but sufficient to be observed with an updated version of that apparatus or with one of the set-ups that, according to the literature, can provide measurements of the Casimir force with 1% (or better) precision.

## 2. Casimir effect with HSMs: previous experimental results

Measurements of the Casimir force using HSMs were reported in [4] (see also [6, 10]).

The set-up used for the experiment is similar to the one originally developed at Bell Laboratories by H B Chan, F Capasso and their collaborators [11, 12] (see figure 2). The force



**Figure 2.** Schematic view of the experimental set-up that has been used for the measurement of the Casimir force between a gold-coated plate and a sphere coated with an HSM (not to scale) [4]. The four top panels illustrate the rotation of the micromachined see-saw in response to the Casimir interaction with the sphere.

sensor consists of a micromachined device that resembles a miniaturized *see-saw*. A polysilicon plate ( $0.5 \times 0.5 \text{ mm}^2$  area), covered with a 5 nm thick chromium adhesion layer and a 200 nm thick gold coating film, is kept in suspension by means of two thin rods that depart from the middle point of two opposite sides of the plate and end onto a post anchored to the substrate. The two rods define the pivot axis of the see-saw. Two electrodes, located underneath the plate in a symmetric configuration with respect to the pivot axis, allow capacitive measurements of the rotation of the top plate with accuracy on the order of  $10^{-7}$  rad, which corresponds to a torque sensitivity of  $\simeq 10^{-15}$  Nm.

The force sensor is mounted on top of a calibrated piezoelectric stage and aligned to a polystyrene sphere ( $100 \mu\text{m}$  radius) in the position illustrated in figure 2. The separation between the sphere and the top plate of the see-saw can be varied by changing the voltage applied to the piezoelectric stage. For separations  $d$  smaller than  $\simeq 400$  nm, the Casimir attraction between the sphere and the plate is sufficient to induce rotations of the see-saw that are larger than the experimental sensitivity. With this set-up, it is thus possible to carry out measurements of the Casimir force between the sphere and the plate from  $d \simeq 400$  nm down to the jump-to-contact point, i.e., to that value of  $d$  where the restoring force of the torsional rods is not sufficient to overcome the attractive force between the two surfaces.

In this experiment, spheres were coated with a Pd-capped Mg-Ni HSM [13]. The mirror was fabricated by means of seven consecutive depositions of Mg-Ni layers, followed by a single deposition of a 5 nm thick palladium cap. Each Mg-Ni layer was obtained by evaporating a 2 nm thick nickel film on top of a 10 nm thick magnesium film. The palladium cap was added to promote hydrogen absorption [9], because, in the conditions of the Casimir force experiment (room temperature and less than atmospheric partial pressure of hydrogen), an uncapped HSM would have not spontaneously absorbed hydrogen. The presence of a metallic layer at the interface

with the external environment had another important function, in that it allowed for the control of electrostatic interactions. In similar uncapped HSMs, the HSM-to-air interface quickly oxidizes. Accumulation of surface charges on the oxide film would have given rise to electrostatic forces that could have easily overcome the Casimir attraction. The deposition of the palladium cap on top of the HSM prevented this process to occur. Furthermore, the presence of a metallic interface, opportunely connected to a power supply, gave the possibility to control the potential difference between the two interacting surfaces, an essential element in all Casimir force experiments [2, 14].

Measurements of the Casimir force  $F_C$  between an HSM-coated sphere and the top plate of the see-saw were carried out as a function of separation  $d$  following the scheme described in [4]. We refer the reader to the original paper for a detailed explanation of the data acquisition technique. Here, we only want to stress that this technique allows one to calibrate the set-up with electrostatic forces and to perform, simultaneously,  $F_C$ -versus- $d$  measurements.

Measurements were carried out in air and in a 4% hydrogen-in-argon mixture at room temperature and nearly atmospheric pressure, and repeated using different spheres and force sensors. A comparison of the data obtained before and after hydrogen loading reveals that there was no significant change in the Casimir force upon hydrogenation of the HSM [4].

### 3. Casimir effect with HSMs: qualitative analysis of the experimental results

The results reviewed above have been qualitatively explained in [4]. The dielectric function of Mg-Ni HSMs is known only in a wavelength region that approximately spans from 0.2 to 2.5  $\mu\text{m}$  [13], where reflectivity switches from  $\simeq 70\%$  to  $\simeq 20\%$  upon hydrogenation. Very little information is available at longer wavelengths, where HSMs might not switch at all. It is thus important to understand whether those longer wavelengths contribute to the Casimir force between surfaces at sub-micrometre separation. If infrared modes are relevant and HSMs do not switch in the infrared spectrum, the decrease of the Casimir force might be too small to be observable with the experimental set-up described in section 2. The following mathematical exercise demonstrates that infrared modes do play a significant role in the Casimir interaction at those distances, and that the change in the force upon hydrogenation is dramatically hampered if one assumes that these HSMs do not switch at long wavelengths.

The Casimir force between a plate made out of a material 1 and a sphere made out of a material 2 separated by a medium 3 can be calculated by plugging the dielectric functions of the interacting surfaces and of the intervening medium directly into the Lifshitz equation for van der Waals interaction [3]:

$$F_C(d) = \frac{\hbar R}{2\pi c^2} \int_0^\infty \int_1^\infty \varepsilon_3 p \xi^2 \{ \log [1 - \Delta_{31}^{(1)} \Delta_{32}^{(1)} e^{-x}] + \log [1 - \Delta_{31}^{(2)} \Delta_{32}^{(2)} e^{-x}] \} dp d\xi \quad (1)$$

where  $\hbar$  is the reduced Planck's constant,  $R$  is the radius of the sphere<sup>2</sup>,  $c$  is the speed of light in vacuum,

$$\Delta_{jk}^{(1)} = \frac{s_k \varepsilon_j - s_j \varepsilon_k}{s_k \varepsilon_j + s_j \varepsilon_k}, \quad \Delta_{jk}^{(2)} = \frac{s_k - s_j}{s_k + s_j} \quad (2)$$

<sup>2</sup> Note that equation (1) relies on the *proximity force approximation*, which is acceptable only if  $R \gg d$ .



and

$$x = \frac{2d\sqrt{\varepsilon_3}\xi p}{c}, \quad s_j = \sqrt{p^2 - 1 + \frac{\varepsilon_j}{\varepsilon_3}}. \quad (3)$$

In equations (1)–(3),  $\varepsilon_j$  stands for the dielectric function evaluated at imaginary frequency,  $\varepsilon_j(i\xi)$ . This quantity is defined by the following relation:

$$\varepsilon_j(i\xi) = 1 + \frac{2}{\pi} \int_0^\infty \frac{\omega \varepsilon_j''(\omega)}{\omega^2 + \xi^2} d\omega, \quad (4)$$

where  $\varepsilon_j''(\omega)$  is the imaginary part of the dielectric function.

In the experiment described in section 2, the intervening medium was either air or a hydrogen-enriched gaseous mixture kept at room temperature and nearly atmospheric pressure. One can thus assume  $\varepsilon_3(i\xi) = 1$  at all frequencies.

Following [4, 6], we can now define an *ad hoc* material whose dielectric function (at real frequencies) is identical to that of gold, with the exception of a wavelength range that spans from  $\lambda_{\min}$  to  $\lambda_{\max}$ , where  $\varepsilon''$  is assumed to be identical to zero. Using equation (1), we can then calculate the Casimir force between a sphere and a plate made out of this hypothetical material. If we assume that infrared modes do not contribute to the Casimir interaction, the values of  $\varepsilon''$  at wavelengths larger than  $\simeq 1 \mu\text{m}$  should not be of any relevance. The Casimir force expected for an *ad hoc* material with  $\lambda_{\min} = 1 \mu\text{m}$  and  $\lambda_{\max} = 200 \mu\text{m}$  should thus not differ from the attraction between two gold surfaces.

Calculations reveal that, in contrast, the value of  $\varepsilon''$  at long wavelengths is of great importance for a correct evaluation of the Casimir force at sub-micrometre separations. For  $d = 100 \text{ nm}$ , for example, the Casimir attraction between a sphere and a plate made out of an *ad hoc* material with  $\lambda_{\min} = 0.3 \mu\text{m}$  and  $\lambda_{\max} = 2.5 \mu\text{m}$  is only  $\simeq 3\%$  smaller than that expected between gold surfaces. However, if one assumes  $\lambda_{\min} = 1 \mu\text{m}$  and  $\lambda_{\max} = 200 \mu\text{m}$ , this difference becomes significantly higher, reaching  $\simeq 35\%$ .

This behaviour is due to the fact that the Lifshitz equation (equation (1)) is intimately connected to the dielectric function of the interacting materials calculated at imaginary frequencies, which can be obtained by means of equation (4). For every imaginary frequency  $i\xi$ , the imaginary part of the complex dielectric function  $\varepsilon''$  is integrated over all real frequencies, with non-negligible contributions arising from a wide range of modes.

In the original paper [4], it was stressed that the theoretical discussion was not accounting for the presence of the thin palladium layer that was added on top of the HSM. The thickness of this coating film is smaller than skin depth and should intuitively not contribute significantly to the interaction. However, the authors suggested that a more rigorous calculation might reveal that this thin metallic layer plays a counterintuitive role. We will come back to this point in section 4.

#### 4. Casimir effect with HSMs: is the modulation of the force observable?

While the arguments reported above provide a reasonable qualitative explanation of the experimental results, it would be interesting to perform a rigorous comparison of the data with theory. Unfortunately, this is not feasible. It is known that the dielectric function of Mg-Ni HSMs strongly depends on their composition and on their hydrogen content upon hydrogenation [13].



Due to the fabrication technique used in the experiment (consecutive deposition of thin layers of magnesium and nickel), it is not possible to establish the exact composition (neither before nor after hydrogenation) of those particular films, which might even have been non-homogeneous. Any attempt to provide a more quantitative comparison of the data with theory would inevitably be based on very rough approximations on the composition of the mirrors. These approximations would lead to ambiguous conclusions.

It is possible, however, to calculate the magnitude of the decrease of the Casimir force between similar HSMs of known composition. The results might provide a clear indication on the feasibility of an experimental observation of the effect, and might also emphasize potential problems that were not considered in the previous experiment.

To achieve this goal, we have calculated the Casimir attraction between a gold plate and a  $100\ \mu\text{m}$  radius sphere coated with a palladium-capped  $\text{Mg}_2\text{Ni}$  mirror. The first step to perform the calculation is to evaluate the dielectric function of the HSM both in its reflective and in its transparent states (i.e., in air and in hydrogen, respectively).

It is known that, in the optical frequency range, HSMs can be well described by a Drude–Lorentz model [15]:

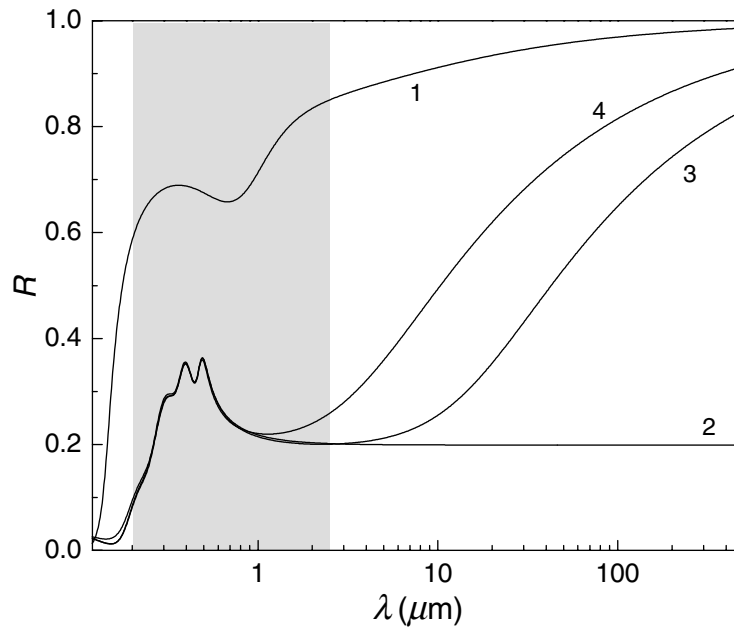
$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\frac{\omega}{\tau}} + \sum_j \frac{f_j}{\omega_{0j}^2 - \omega^2 - i\beta_j\omega}. \quad (5)$$

The model includes a Drude contribution arising from the presence of free charge carriers with plasma frequency  $\omega_p$  and relaxation time  $\tau$ , and a series of Lorentz oscillators with strengths  $f_j$ , resonance frequencies  $\omega_{0j}$  and damping terms  $\beta_j$ . Because equation (5) is analytic, the dielectric function at imaginary frequency can be simply obtained by substituting  $\omega$  with  $i\xi$ :

$$\varepsilon(i\xi) = \varepsilon_\infty + \frac{\omega_p^2}{\xi^2 + (\xi/\tau)} + \sum_j \frac{f_j}{\omega_{0j}^2 + \xi^2 + \beta_j\xi}. \quad (6)$$

The Drude–Lorentz parameters  $\omega_p$ ,  $\tau$ ,  $f_j$ ,  $\omega_{0j}$ ,  $\beta_j$  and  $\varepsilon_\infty$  for  $\text{Mg}_2\text{Ni}$  HSMs were obtained by Lohstroh *et al* [16, 17], who analysed reflectivity and transmittivity measurements in the wavelength region spanning from ultraviolet to near infrared. According to their results, for mirrors in the reflective state (i.e., mirrors in air before exposure to hydrogen), one Lorentz oscillator and one Drude term are sufficient to reproduce the experimental data. For mirrors in the fully loaded transparent state (i.e., after complete hydrogenation, corresponding to  $\text{Mg}_2\text{NiH}_{4-\delta}$ , with  $\delta \simeq 0$ ), four Lorentz oscillators are necessary, but no Drude tail is needed<sup>3</sup>. Optical data, in fact, do not reveal any Drude-like behaviour up to the longer wavelengths investigated ( $\simeq 2.5\ \mu\text{m}$ ). To understand the relevance of this point, in figures 3 and 4 we plot, respectively, the reflectivity and the dielectric function of  $\text{Mg}_2\text{Ni}$  HSMs before and after switching. The shaded area represents the wavelength region where experimental data are available. Curve 1 refers to the fitting function proposed by Lohstroh *et al* for  $\text{Mg}_2\text{Ni}$  HSMs before hydrogenation. Curve 2 corresponds to the fitting function proposed by the same group for the hydrogenated state, with

<sup>3</sup> We thank Lohstroh *et al* for providing us with the values of the Lorentz parameters:  $\varepsilon_\infty = 2.45$ ,  $\sqrt{f_1} = 2.67\ \text{eV}$ ,  $\omega_{01} = 2.54\ \text{eV}$ ,  $\beta_1 = 0.40\ \text{eV}$ ,  $\sqrt{f_2} = 3.62\ \text{eV}$ ,  $\omega_{02} = 3.08\ \text{eV}$ ,  $\beta_2 = 0.69\ \text{eV}$ ,  $\sqrt{f_3} = 4.32\ \text{eV}$ ,  $\omega_{03} = 3.87\ \text{eV}$ ,  $\beta_3 = 1.40\ \text{eV}$ ,  $\sqrt{f_4} = 4.48\ \text{eV}$ ,  $\omega_{04} = 5.64\ \text{eV}$  and  $\beta_4 = 3.03\ \text{eV}$ .



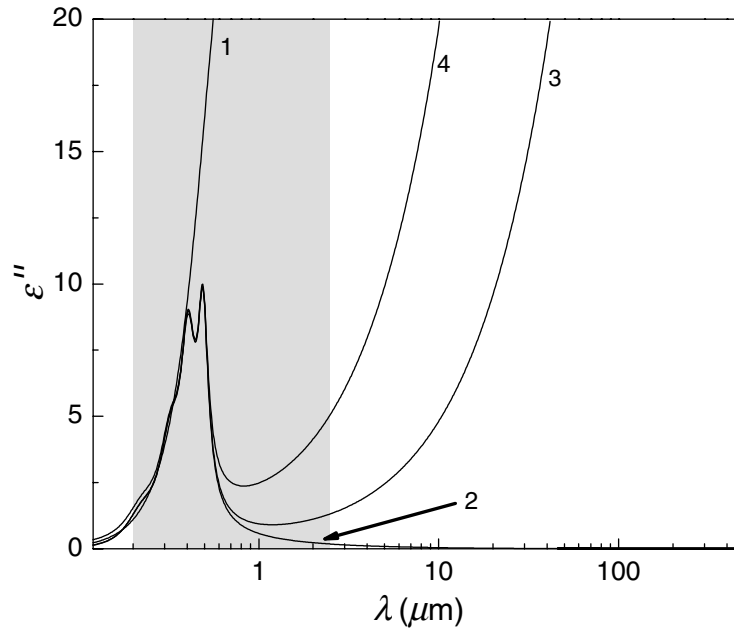
**Figure 3.** Reflectivity of  $\text{Mg}_2\text{Ni}$  HSMs as a function of wavelength. The shaded area represents the wavelength region where experimental data are available. Curve 1 refers to a Drude–Lorentz model that fits the optical data of the mirror in its reflective state (before switching). Curves 2, 3 and 4 refer to hydrogenated HSMs. Curve 2 represents a Lorentz model obtained by fitting the optical data. Curve 3 is equivalent to curve 2, with the addition of a Drude term obtained from the combined analysis of Hall effect and resistivity measurements. Curve 4 is the sum of curve 2 and a Drude term with *ad hoc* plasma frequency and relaxation time.

four Lorentz oscillators and no Drude term. Curves 3 and 4 were obtained by us using the same function as in curve 2 with an additional Drude tail<sup>4</sup>. The precise meaning of these curves will be clarified later in the text. Here we want to stress that curves 2 and 3, which have different behaviour in the infrared, are very similar over the whole wavelength region where experimental data are available. None of these curves is thus in contradiction with the experiment. It is then evident that a correct extrapolation of the dielectric function at longer wavelengths cannot rely on these data. Unfortunately, these data still represent the only direct source of information available: to our knowledge, in fact, optical properties of HSMs have never been systematically measured beyond  $\simeq 2.5 \mu\text{m}$ <sup>5</sup>.

In the absence of more direct experiments, it is still possible to estimate the Drude term in fully hydrogenated  $\text{Mg}_2\text{Ni}$  HSMs by a combined analysis of Hall effect and resistivity

<sup>4</sup> For curve 4, the oscillator strengths of the Drude–Lorentz model  $f_j$  were multiplied by a factor of 0.9.

<sup>5</sup> In [18], the authors reported measurements of the reflection and transmission coefficients up to  $6 \mu\text{m}$ . However, raw data were not analysed in terms of the Drude–Lorentz model, and cannot be used in our calculations. It is however important to emphasize that those measurements do not reveal any Drude tail in the wavelength region between  $2.5$  and  $6 \mu\text{m}$ , still in agreement with curves 2 and 3.



**Figure 4.** Imaginary dielectric function of  $\text{Mg}_2\text{Ni}$  HSMs as a function of wavelength. Symbols are equivalent to those used in figure 3.

measurements. Hall effect experiments provide a direct measurement of the density of free charge carriers  $n$  [19]. If resistivity is known, one can then calculate the dissipation factor and the plasma frequency of the Drude term using the following equations [19]:

$$\tau = \left( \frac{0.22}{\rho_\mu} \right) \left( \frac{r_s}{a_0} \right)^3 \times 10^{-14} \text{ s}, \quad (7)$$

$$\omega_p = 7.27 \left( \frac{a_0}{r_s} \right)^{3/2} \times 10^{16} \text{ rad s}^{-1}, \quad (8)$$

where  $\rho_\mu$  is the resistivity in  $\mu\Omega \text{ cm}$ ,  $a_0$  is the Bohr radius and  $r_s$  is given by

$$r_s = \left( \frac{3}{4\pi n} \right)^{1/3}. \quad (9)$$

Magnetoresistance and charge carrier density of  $\text{Mg}_2\text{Ni}$  HSMs were measured by Enache *et al* [20] for different hydrogen concentrations. For fully loaded mirrors, the authors obtained  $n = 9.28 \times 10^{20} \text{ carriers cm}^{-3}$  and  $\rho = 12.9 \text{ m}\Omega \text{ cm}$ .

We can now comment further on figures 3 and 4. We recall that curve 1 represents the HSM in its high reflectivity state<sup>6</sup>, and that curve 2 was obtained by fitting the optical data

<sup>6</sup> It is interesting to note that, for the reflective state ( $\text{Mg}_2\text{Ni}$ ), the combined analysis of Hall effect and resistivity measurements gives rise to plasma frequency and dissipation factor values that are in agreement with those obtained from optical data [16].

from visible to near infrared with four Lorentz oscillators and no Drude term. This model, which is equivalent to assume that HSMs become highly transparent at long wavelengths, is not consistent with Hall effect and resistivity measurements, and must thus be considered incorrect in the infrared spectrum. Curve 3 is equivalent to curve 2 plus a Drude term calculated according to the arguments reported above; it represents the best estimation of reflectivity and dielectric function of  $\text{Mg}_2\text{NiH}_{4-\delta}$  (with  $\delta \simeq 0$ ) that one can obtain with the data available in literature. Curve 4 is again an *ad hoc* model that will serve to stress once more the importance of infrared modes in the Casimir interaction. The properties of this hypothetical mirror are in contradiction with Hall effect and resistivity measurements.

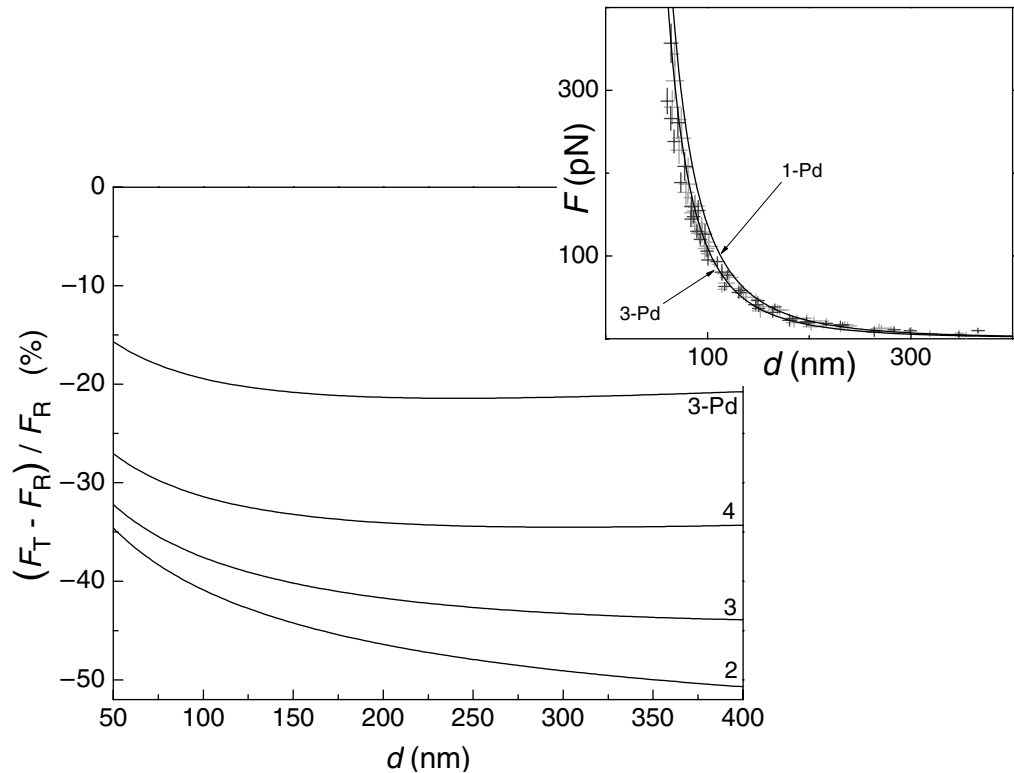
At this point, we have enough information to model the  $\text{Mg}_2\text{Ni}$  mirrors in air (model 1) and in hydrogen (model 3), and to discuss the consequences that the use of erroneous models for the hydrogenated state would have in the calculation of the Casimir force upon hydrogenation (models 2 and 4). The presence of a thin palladium layer deposited on top of the mirror can be introduced in the calculation by means of a model developed by Parsegian and Ninham [21], according to which the Casimir force between a material 2 and a material 1 covered with a cap layer 4 of thickness  $t_4$ , immersed in a medium 3, is obtained from equation (1), replacing  $\Delta_{31}^{(1,2)}$  with

$$\Delta_{31}^{(1,2)} \rightarrow \frac{\Delta_{34}^{(1,2)} + \Delta_{41}^{(1,2)} e^{-(xt_4s_4/pd)}}{1 + \Delta_{34}^{(1,2)} \Delta_{41}^{(1,2)} e^{-(xt_4s_4/pd)}}. \quad (10)$$

In order to run the calculation, it is still necessary to evaluate the dielectric function of the gold plate and of the palladium layer. For these materials, we use the tabulated data for the corresponding bulk metal [22] plus a Drude term as described in [23] (for gold) and [24] (for palladium).

In figure 5, we report the results of the calculations. The graphs show the decrease of the Casimir force between a gold plate and a  $100 \mu\text{m}$  sphere coated with a  $\text{Mg}_2\text{Ni}$  HSM, alternatively exposed to either air or hydrogen.  $F_R$  and  $F_T$  indicate the force for the reflective state and for the optically transparent hydrogenated state, respectively.  $d$  is the separation between the sphere and the plate. Different curves refer to different models of the hydrogenated state (see table 1), and have been numbered in accordance with figures 3 and 4. Curves 2, 3 and 4 were obtained neglecting the presence of the palladium film. Concerning curve 3-Pd, a 5 nm thick palladium layer was added to the calculation of both  $F_T$  and  $F_R$ . From this graph, two important conclusions can be drawn.

- Curves obtained with no Drude term (curve 2), with the correct Drude term (curve 3) and with the fictitious Drude term (curve 4) are significantly different. This result emphasizes once more that if one assumes that hydrogenated HSMs remain highly reflective at long wavelengths, the decrease in the force upon hydrogenation is much smaller than that expected with a wide band switching mirror. Long wavelengths are thus indeed relevant, as already pointed out in [4].
- The 5 nm thick palladium capping layer deposited on top of the HSM plays a dramatic role in the interaction. At 100 nm separation, the change of the Casimir force upon hydrogenation for an uncapped mirror is larger than 35% (curve 3); if the palladium layer is added (curve 3-Pd), the effect reduces to  $\simeq 20\%$ . Further calculations show that a 10 nm thick palladium would decrease the change of the force at 100 nm to  $\simeq 10\%$ . The palladium layer represents a serious obstacle for the observation of large Casimir force modulations.



**Figure 5.** Expected decrease in the Casimir force between a gold plate and a  $100 \mu\text{m}$  radius sphere coated with an HSM exposed to either air or hydrogen.  $F_R$  and  $F_T$  indicate the force for the reflective state and for the transparent hydrogenated state, respectively.  $d$  is the separation between the sphere and the plate. Calculations were performed using the dielectric functions reported in figure 4 (see also table 1). Different curves refer to different models of the hydrogenated state, and have been numbered in accordance with figures 3 and 4. The suffix ‘Pd’ indicates whether the 5 nm thick palladium coating layer was taken into account in the calculations or not. Inset: comparison of the results of the calculations with the experimental data reported in [4]. Light and dark grey symbols refer to measurements performed in air and hydrogen, respectively.

In the inset of figure 5, we report a direct comparison of the calculations with the experimental data of [4]. We want to stress once more that the curves represent the results of calculations performed using the properties of materials that probably have a different composition with respect to the ones used in the experiment. Nevertheless, the graph allows us to emphasize that, even in the *best case scenario* of fully loaded HSMs with fairly good switching properties in the infrared, the effect could have been hardly observed with the experimental sensitivity of the set-up described in [4].

It is to be noted that our calculation relies on the hypothesis that the HSM in use undergoes full hydrogen loading upon hydrogenation. Partial loading would significantly alter the result. It is known, in fact, that the loading process in  $\text{Mg}_2\text{Ni}$  HSMs (and, more generally, in most of Mg-Ni compounds) involves a remarkable self-organized double layering [25]. Hydrogen first

**Table 1.** Summary of the models used for the calculations reported in the text. The asterisk added in the description of model 4 indicates that, for this particular model, the oscillator strengths of the Lorentz term have been multiplied by a factor of 0.9.

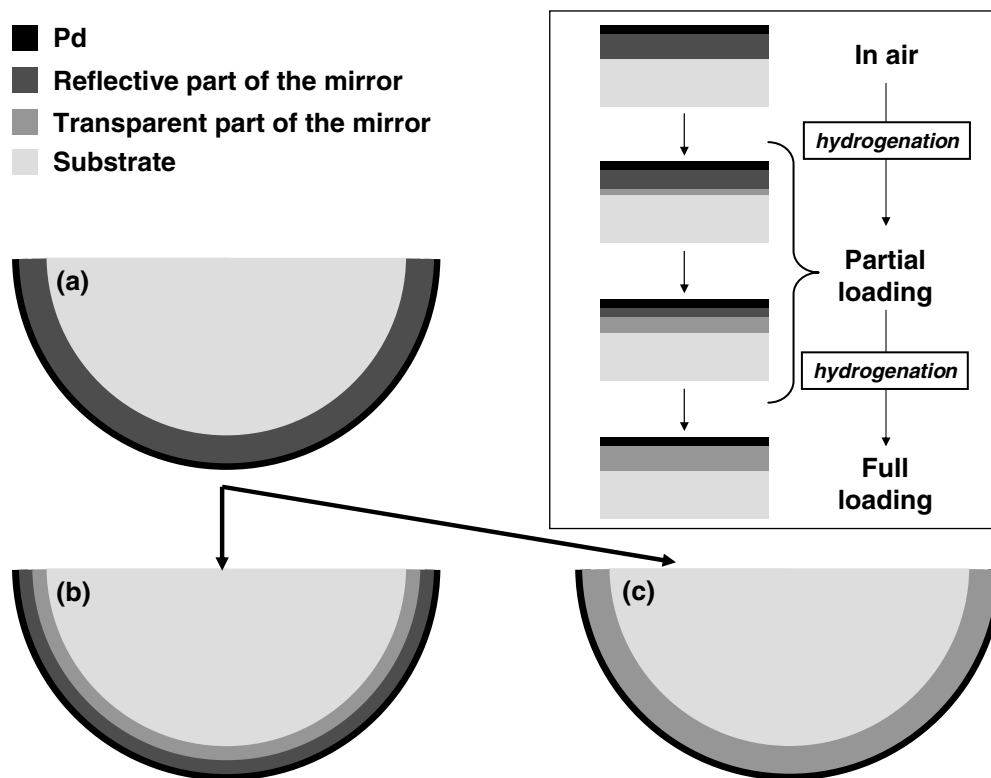
Material	Description
HSM reflective state Model 1	1 Lorentz oscillator [16] plus Drude term: $\omega_p = 1.24 \times 10^{16} \text{ rad s}^{-1}$ ; $\tau = 9.9 \times 10^{-16} \text{ s}$
HSM hydrogenated state Model 2	4 Lorentz oscillators [17] No Drude term
HSM hydrogenated state Model 3	4 Lorentz oscillators [17] plus Drude term: $\omega_p = 0.17 \times 10^{16} \text{ rad s}^{-1}$ ; $\tau = 2.96 \times 10^{-16} \text{ s}$
HSM hydrogenated state Model 4	4 Lorentz oscillators [17]* plus Drude term: $\omega_p = 1.43 \times 10^{16} \text{ rad s}^{-1}$ ; $\tau = 1.82 \times 10^{-17} \text{ s}$
Gold surface	Tabulated data [22] plus Drude term: $\omega_p = 1.37 \times 10^{16} \text{ rad s}^{-1}$ ; $\tau = 1.88 \times 10^{-14} \text{ s}$ [23]
Palladium layer	Tabulated data [22] plus Drude term: $\omega_p = 0.83 \times 10^{16} \text{ rad s}^{-1}$ ; $\tau = 4.28 \times 10^{-14} \text{ s}$ [24]

penetrates inside the film and quickly reaches the substrate, from where hydrogenation starts (see inset of figure 6). Partial loading leads to a bi-layered structure formed by a transparent layer close to the substrate and a non-switched metallic layer close to the palladium film. This non-switched metallic layer is still highly reflective. Its presence would have a dramatic effect on the Casimir force, as it increases the overall thickness of the metallic film above the hydrogenated part of the mirror (see figure 6). Note that it is not possible to establish whether the HSMs used in the experiment of [4] were fully loaded. The presence of a non-switched layer below the palladium film might have contributed to reduction of the magnitude of the Casimir force difference between measurements in air and in hydrogen.

Our analysis also neglects the correction to the Casimir force due to the surface roughness of the interacting objects. These corrections cannot be explicitly calculated *a priori*, because the morphology of HSMs strongly depends on the deposition technique [26]. In this respect, it is worth stressing that the surface profile of HSMs in air undergoes large variations during the first few hydrogen loading and unloading cycles [26, 27], after which no more morphological adjustments are observed. Therefore, reproducible modulations of the Casimir force with HSMs can be achieved only after the mirrors have been switched a few times. It is also evident that for a correct interpretation of the data, accurate measurements of the morphology of the surfaces both in air and in hydrogen are needed, because the alteration of the roughness upon hydrogenation could mimic or hinder the decrease of the Casimir force induced by the change of the optical properties of the mirrors.

## 5. Conclusions

Tuning the Casimir force with HSMs is a fascinating opportunity that calls for deeper investigations. The only measurements reported so far [4] were affected by a relatively large



**Figure 6.** Schematic view showing that partial loading of HSMs plays a detrimental role in Casimir force modulation experiments. Sketch (a) refers to the mirror before hydrogenation; sketch (b) shows the layered structure that would form in case of partial hydrogen loading, with a relatively thick reflective layer still present at the interface; sketch (c) illustrates the absence of layered structures at the interface in fully loaded mirrors. Inset: schematic diagram of the hydrogen loading mechanism in Mg-Ni HSMs.

experimental error, which was mainly due to the fact that the experiment was carried out at atmospheric pressure. The change in the force upon hydrogenation was smaller than intuitively expected, and was not sufficient to be observed. A qualitative theoretical analysis first showed that the results could have been explained assuming that the dielectric function of HSMs switches only in a limited wavelength region that spans from ultraviolet to near infrared, where optical data are available [4]. Longer wavelengths play in fact an important role in the interaction: if the mirror remains highly reflective in the infrared part of the spectrum, the change in the force upon hydrogenation can be smaller than the experimental sensitivity. A more complete theoretical analysis of the experiment was not performed, because the exact composition of the mirrors was not known.

In this paper, we have calculated the expected decrease of the force for similar mirrors of known composition ( $\text{Mg}_2\text{Ni}$ ). The results confirm the main conclusion of [4]: for an accurate comparison of experimental data with theory, it is necessary to know the value of the dielectric functions of the interacting materials over a broad wavelength range that extends up to the far infrared region. Furthermore, the presence of a palladium capping layer strongly reduces the effect. Still, at 100 nm separation, the Casimir force is expected to decrease  $\simeq 20\%$  upon



full hydrogenation. We thus believe that it is possible to design a new experiment for the first observation of an *in situ* modulation of the Casimir force between HSMs. A significant reduction of the experimental uncertainty can be obtained by performing the measurements in vacuum. It is known that, in vacuum, hydrogenated HSMs do not release hydrogen. One could thus load or unload the mirror by flushing the measuring chamber with proper gases at atmospheric pressure (hydrogen for loading, and air for unloading), and then pump vacuum during Casimir force measurements. In order to obtain a more pronounced effect, it would be also desirable to reduce the contribution of the thin palladium layer deposited on top of the HSM. The use of films thinner than 5 nm is in principle a possible solution to this problem. However, the resulting layer would probably be discontinuous after switching [27] and would not allow a complete control of electrostatic forces. A possible alternative is to replace palladium with a metal that wets the mirror better than palladium does, and then use hydrogen diffusion from a palladium pad located away from the interaction region (i.e., away from the portion of the sphere that, upon assembling of the experimental set-up, stands in front of the force sensor) to obtain uniform switching [28, 29]. This choice might allow one to reduce the thickness of the cap layer down to 2 or 3 nm. A more precise experiment would also probably stimulate a more accurate comparison with theory. Direct measurements of the dielectric functions in the infrared wavelength region and of the surface morphology of the films deposited onto the interacting surfaces would certainly facilitate a rigorous interpretation of the experimental data.

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