Casimir forces and phase-change materials

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ABSTRACT

Phase-change materials are well known for their structural transformation producing a significant change in the optical properties and this is exploited in optical data storage systems. Optical properties are also the defining factor for the so-called Casimir force, that is related to the ground-state of the electromagnetic field in vacuum. We demonstrate that for a single material system a significant variation in the Casimir force can be achieved. Changes in the force of up to 20% at separations of ~100 nm between Au and AgInSbTe (AIST) surfaces were found (in both experiment and theory) by changing the AIST from the amorphous to the crystalline phase. The present finding paves the way to force control in nano-systems, such as micro- or nano-switches. However, then the effect of a capping layer on the phase-change film has to be included. We show here that SiO_2 capping layers significantly reduce the Casimir force contrast between the amorphous and crystalline phase and therefore they should be applied as thin as possible.

Key words: Casimir force, AgInSbTe, amorphous films, crystalline films, ellipsometry, atomic force microscopy

1. INTRODUCTION

Casimir forces [1-8] arise between two surfaces due to the quantum zero-point energy of the electromagnetic field. The surfaces restrict the allowed wavelengths and thus the number of field modes within the cavity, which locally depresses the zero-point energy of the electromagnetic field (see Fig.1). The reduction depends on the separation between the plates; thus there is a force between them, which for normal materials is always attractive [1]. The zero point energy manifests itself as quantum fluctuations, which in the small separation limit give rise to the familiar van-der-Waals force. The original calculation of the Casimir force assumed two parallel plates with an infinite conductivity [1]. This was later modified to include the dielectric properties of real materials and the intervening medium [2,3], providing the first glimpse of possible methods to control the magnitude and even the direction of the force. This finding has motivated our attempts to manipulate the dielectric properties of a material and hence generate force contrast [9-11]. A particularly exciting possibility is to produce a 'switchable' force by employing materials whose optical properties can be changed *in situ* in response to a simple stimulus [9,10]. So far the only significant contrast that has been demonstrated is between different materials [11]. To obtain a large Casimir force contrast for a single material requires a large modification of its dielectric response, which has not been achieved in materials used up to now.
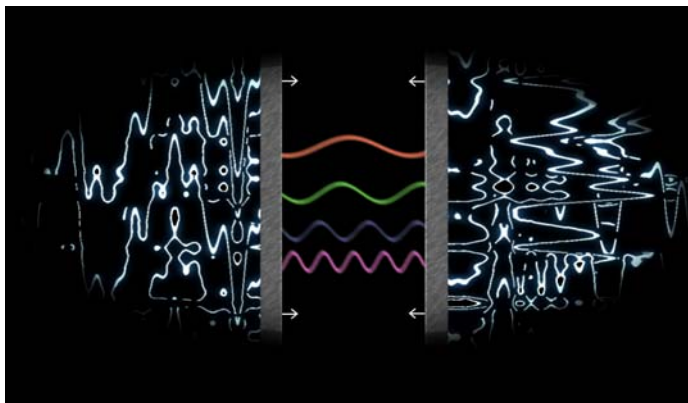

Figure 1. *Impression of the Casimir force as a consequence of vacuum fluctuations of the electromagnetic field. Outside the cavity there is a 'fluctuating sea' of virtual photons or fields, inside only certain modes of these fields can exist due to the imposed boundary (where the field must be zero). The 'pressure' of the 'sea' outside is bigger than the pressure due to the fields inside and it will result in an attractive force between the opposite walls of the cavity.*

Here we demonstrate that phase-change materials (PCMs) [12-21], that is, materials that are renowned to switch between an amorphous and crystalline phase, are very promising candidates to achieve a significant force contrast without changing their composition. These materials are already used in rewriteable optical data storage [13,14,23-25], where the pronounced optical contrast between the amorphous and crystalline state is employed to store information.

2. EXPERIMENTS

In order to measure Casimir forces with PCMs, we prepared 1 μm thick amorphous AgInSbTe (AIST) thin films onto standard Al coated Si wafers, of which half of the AIST films were annealed to the crystalline state. The samples were optically characterized by ellipsometry in the frequency range ω=0.04-8.9 eV.

The Casimir force measurements were performed using the dynamic AFM mode within an ultra-high vacuum (UHV) Atomic Force Microscope (Omicron VT STM/AFM) [27, 28]. Forces were measured in the sphere-plate geometry between a (100 nm thick) Au-coated sphere 20.2 μm in diameter, attached to the end of a cantilever. The latter initially vibrates at its resonant frequency, 83.6 kHz, far from the surface. As the sphere approaches the PCM surface, we measure the frequency shift induced by the sphere-plate interaction, which is proportional to the force gradient in the linear approximation. Each experimental force curve is an average of 13 measurements taken in different areas on both samples. The force measurement method and the experimental set-up are described in detail in [28].

3. RESULTS & DISCUSSION

Ellipsometry results in the frequency range ω=0.04-8.9 eV are shown for the amorphous and crystalline AgInSbTe (AIST) thin films (onto standard Al coated Si wafers) in Fig. 2. For the crystalline sample the measurements were directly inverted to obtain the dielectric function [22]. For the amorphous film, because it is transparent in the infrared (IR) range, the system was modelled as an amorphous film above an optically thick Al substrate. The substrate optical properties are important only in IR range, where absorption of the film is very weak. Therefore, it is justified to use tabulated data for the Al substrate.
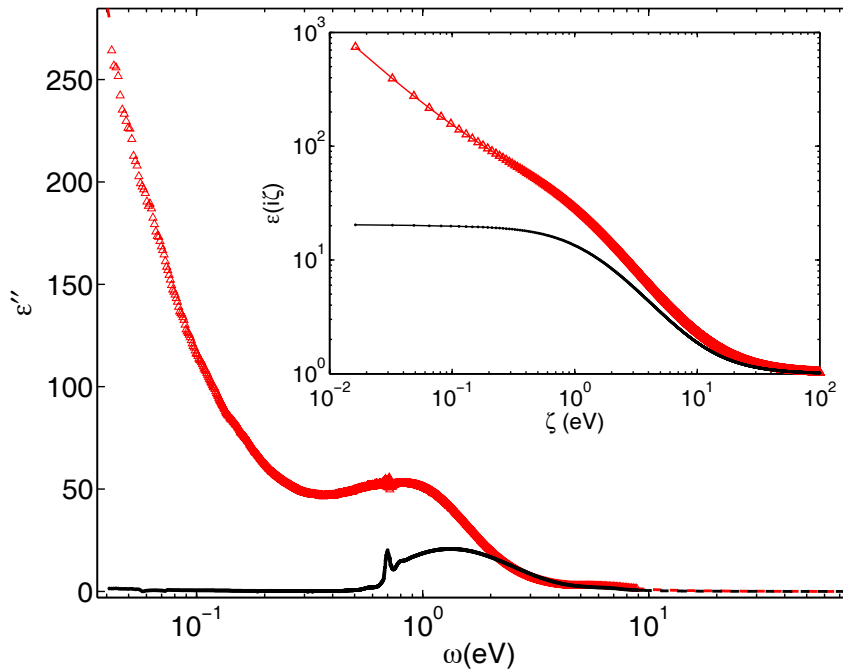

Figure 2. Absorptive part of the dielectric function for the crystalline (red) and amorphous (black) state of the AIST film obtained with ellipsometry as a function of frequency. The inset shows the same dielectric functions at imaginary frequencies ζ, which are necessary for the Casimir force calculations using Lifshitz theory.

Since the crystalline film exhibits metallic conductivity, a Drude model was fitted to the measured IR data enabling extrapolation below $\omega<0.04$ eV, where data are not available. For the amorphous state this range has an insignificant effect on the force. At high frequencies $\omega>8.9$ eV, where absorption is already small, the imaginary part of the dielectric function $\varepsilon(\omega) = \varepsilon'(\omega) + j\,\varepsilon''(\omega)$ was extrapolated as $\sim 1/\omega^3$. The extrapolations are justified by a good Kramers-Kronig (KK) consistency for amorphous and crystalline films, and good agreement with the permittivities of the films found previously [23]. As can be seen from Fig. 2, the transformation from the amorphous to the crystalline state leads to drastic changes of the optical properties. These pronounced changes have been recently attributed to a change of bonding upon crystallization [13,14,23]. The large change of the dielectric function upon crystallization suggests that a significant change in the Casimir force should be observed.

The measured dielectric response allows Casimir force calculations using the so-called Lifshitz theory (Fig. 3) [2,3], for which the force depends on the dielectric function at imaginary frequencies (inset Fig. 2). However, such forces are also affected by the surface roughness. The typical RMS roughness of the samples was a few nm, but with a few isolated local peaks as evidenced by atomic force microscopy (AFM) analysis (lower inset in Fig.3). This small roughness is negligible for the Casimir force calculation at separations above 70 nm [26]
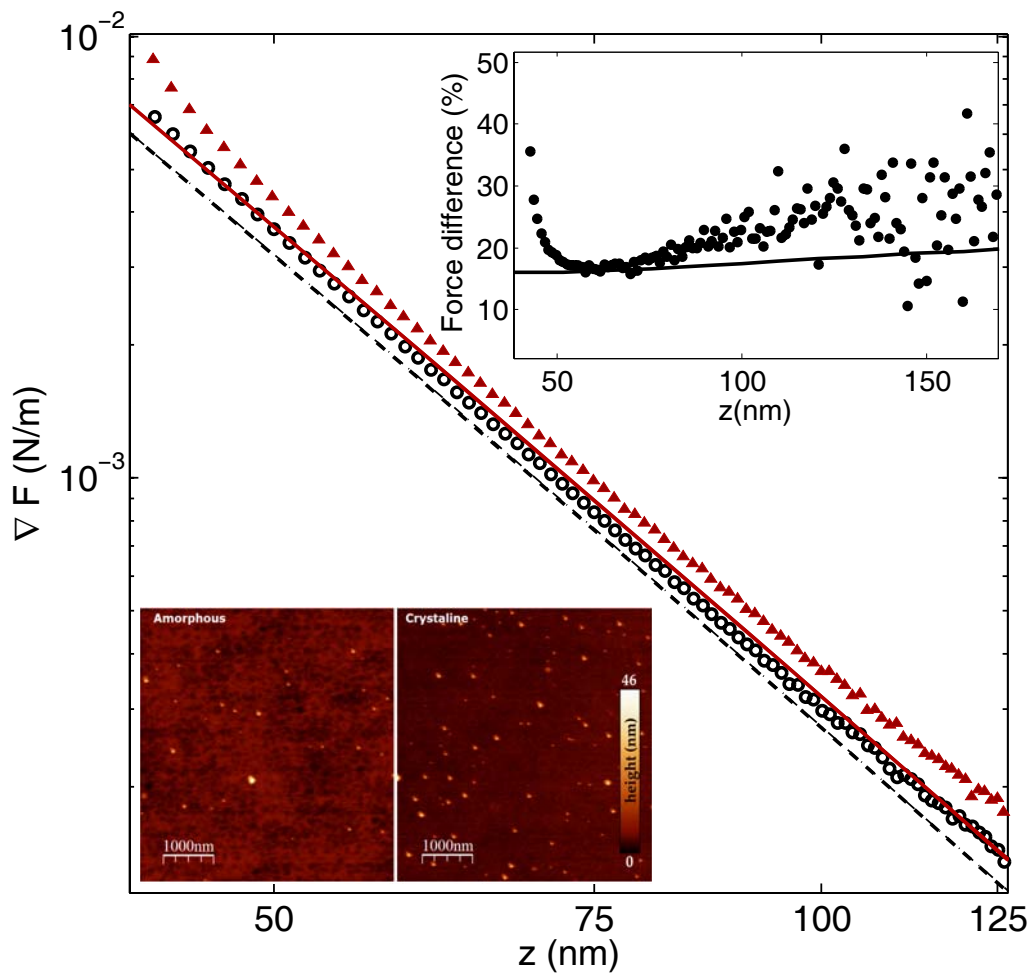

Figure 3. *Casimir force gradient measurement for the crystalline (red Δ) and amorphous (•) phase. The theory curves are indicated with lines (red —) crystalline, and (−· −) amorphous. The upper inset shows the relative difference between the two force states, normalized with respect the amorphous state, for both the experimental (•) and theoretical (—) data . The lower inset shows an AFM topography of amorphous (left) and crystalline (right) films.*

Results of Casimir force measurements are shown in Fig.3. Precise comparison of force measurements with theory is only possible if we experimentally determine (electrostatically) several, *a priori* unknown, parameters such as the starting separation distance Z_0 for the force measurement (corresponding here to the shortest separation), the cantilever spring constant k, and the contact potential difference V_0 [28]. The calibration is performed by measuring the force gradient versus separation distance for two different applied bias voltages V_b on the sphere yielding a gap voltage $\Delta V = V_b - V_0$. The contact potential V_0 may not be constant [11,27,29] but instead can depend on the separation distance Z between sphere and sample surface. Prior to force acquisition, the determination of V_0 is performed at only one distance $Z_0 = 42.8 \pm 0.5$ nm for the amorphous, and $Z_0 = 42.9 \pm 0.4$ nm for the crystalline phase sample. Then we define $V_0 = 0$ at $Z = Z_0$ as the reference potential, and the two values are chosen for V_b (-0.5 V, +0.5 V) to obtain the electrostatic force curves. Determination of Z_0 and k is achieved by fitting the average of these two force measurements after subtraction of the Casimir contribution (measured for $V_b = 0$ V), without the calibration being affected by variations in V_0 [28]. The fit gives consistent spring constants, namely, $k = 10.8 \pm 0.3$ N/m for the amorphous film, and $k = 10.7 \pm 0.3$ N/m for the crystalline film.

The experimental uncertainty in the force measurement as deduced from the standard deviation of the cantilever spring constant k and the starting separation distance Z_0 is about 7% for both samples. Therefore, the upper inset in Fig.3 demonstrates unambiguously that the gradient of the Casimir force increases in magnitude by approximately 20 % as a result of the transition from the amorphous to the crystalline state. Both the size and the sign of this force change upon crystallization are in excellent agreement with the theoretical calculations. At short separations (< 55 nm) the increase in the difference can probably be attributed to the larger roughness of the crystalline state (lower inset Fig.3) leading to a larger force [26].

Although experiment and theory agree well with respect to the *difference* in force between the amorphous and crystalline states, the theory does not predict the *absolute* values of the forces for the two states. The theory based on the measured optical properties predicts a force smaller than the measured one by 8-18 %. The deviation is smaller for the amorphous sample but in both cases it is larger than the experimental and theoretical errors. This deviation cannot be explained by a vertical drift of the AFM probe since the feedback loop maintains the sphere at separation Z_0 from the surface (positioning accuracy better than ~0.1 nm). In addition, it cannot be explained by the fact that the electrostatics have been performed using an approximate formula for capacitance gradient [28] which leads to an error of Z_0 of ~0.2 nm. Also, in order to check the force measurements we used a sample coated with low RMS roughness Au (~1 nm) and a close agreement was found between the measured and theoretically predicted forces. Possible uncertainties in the optical properties of PCM due to low and high frequency extrapolations, variation of the substrate properties or film thickness are also excluded since they have small influence on the force calculation.

Hence, the observed deviation between theory and experiment can be attributed to surface roughness as discussed recently in [30]. Indeed, the electrostatic force involves a larger interaction area on the plate than the Casimir force [30]. Larger areas contain more high peaks so that the averaged surface of the plate will be located higher than for smaller areas [30]. This is specific to the PCM roughness as the inset in Fig. 3 shows. As a result the absolute separation as determined from the electrostatic calibration underestimates the separation in the case of the Casimir interaction. This difference can be ~1-2 nm [30], and it is smaller for the amorphous film. In fact, if the experimental force data are shifted to the left by 1-2 nm, the agreement with the theory is nicely restored.

The measured contact potential difference between the gold-crystalline and the gold-amorphous case is about 23 mV (Fig. 4). A variation of the potential with distance is also observed, which is the same for both phases, and it therefore does not generate a force contrast. Hence, these results show that electrostatic forces do not overshadow the Casimir force contrast. For further details about electrostatics we refer to Refs. [28,31].
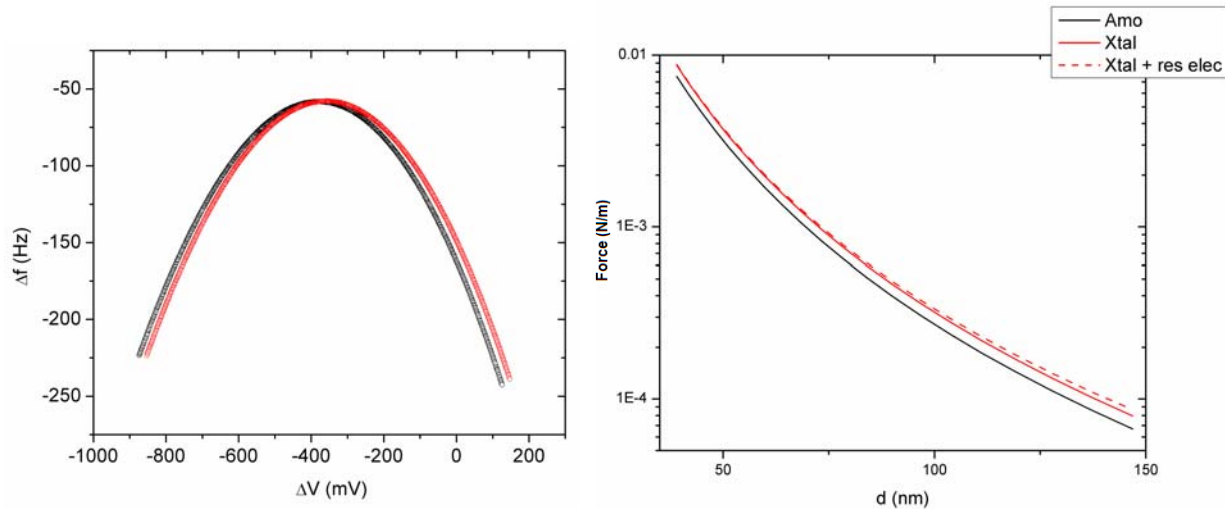
Figure 4. *Left: Frequency shift of the resonator versus applied potential. This yields a parabola with a maximum when the contact potential between gold and AIST is minimized. The contact potential difference for the amorphous and crystalline phases is 23 mV. The effect of this residual difference is small compared to the total force gradient (N/m) for separations d < 150 nm, implying that in this range the Casimir force contrast dominates; see right figure.*

For *in situ* switching phase-change materials, protective capping layers are required that are generally transparent and non conductive like SiO_x. For this reason we show calculations with Lifshitz theory [2,3] for AIST with 5 and 10 nm silica (SiO_2) protective layers (Fig. 5). These capping layers decrease the force contrast with 5-10% (Fig. 5). On the other hand such non conductive protective layers will also contribute to electrostatic charging, and this remains to be investigated. However if the charging of the system does not change much for the two states, then Casimir force contrast may still dominate. At this point AIST is the only PCM that has been investigated for Casimir force contrast, thus there may still be room for improvement of the force contrast with a different PCM.
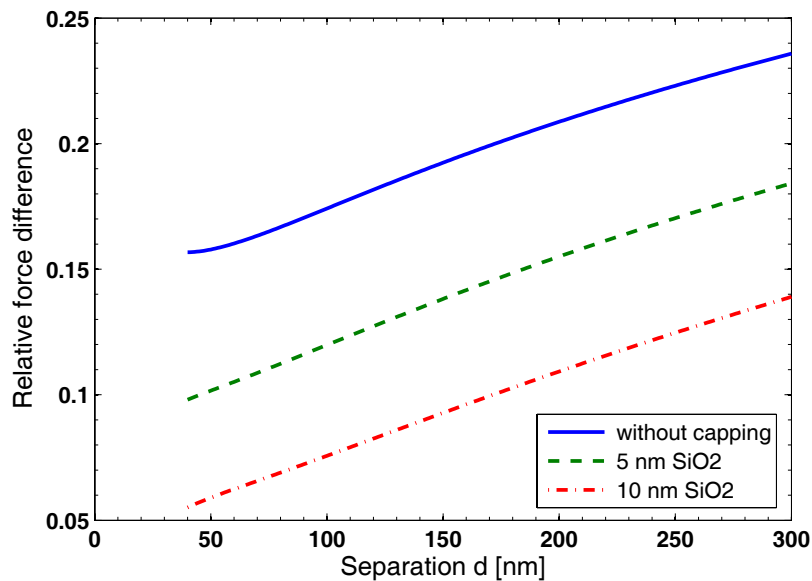

Figure 5. *Calculated Casimir force contrast (F_{cryst} - F_{amorph})/F_{amorph}. Force contrast is shown for bare AIST, and for AIST with a 5 nm and 10 nm silica protective layers. A 5 nm capping layer decreases the contrast with ~5%.*

4. CONCLUSIONS

As expected from the pronounced difference in the dielectric functions of the amorphous and crystalline phases in phase-change materials, a significant difference in the Casimir force between the PCM and Au is found for the two phases. The measured force contrast, about 20% at a PCM-Au separation of 100 nm, is the largest reported to date for a switchable material [9,10]. These experimental results agree well with theory. Although switching a large area of PCM requires high currents, when the nanometer regime is entered modest currents are sufficient to switch the PCM material. We investigated the effect of SiO_2 capping layers and found that they significantly reduce the Casimir contrast and therefore have to be applied with caution. Nevertheless, the fast switching, good scalability down to the nanometer regime [24], large dielectric contrast and large Casimir force contrast deem PCMs to be promising, if not the only, candidates for a switchable Casimir force device.

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