11 Modulation of optical properties and Casimir forces

11.1 Introduction
The idea of switching the Casimir force with materials that can be optically modulated is not new [1]. Yet it is difficult to find switchable materials with a sufficiently large optical contrast to significantly affect the Casimir force. Since our group was working a lot with Phase Change Materials (PCMs), at some point the idea emerged that these materials could be very well suited for this purpose. While we could do sample characterization and theory, our force measurement device did not have sufficient precision to detect this effect. Therefore, together with prof. C. Binns and G. Torricelli, who had a similar idea and who had developed a much better Ultra High Vacuum force measurement device for this cause, and prof. M. Wuttig who delivered specialized samples, we demonstrate here a controllable variation in the Casimir force. Changes in the force of up to 20% at separations of ~100 nm between Au and AgInSbTe (AIST) surfaces were achieved by switching the AIST films between the amorphous and crystalline states. PCMs are well known for this structural transformation, which are exploited in DVD's and Phase Change Random Access memory.

11.2 Samples & Ellipsometry
So far significant Casimir force contrast has only been demonstrated between different materials [2]. 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. Here we demonstrate that phase change materials (PCMs) [3-12], that is, materials that 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 [4, 5, 14-16], where the pronounced optical contrast between the amorphous and crystalline state is employed to store information. Therefore our study differs significantly from the previous attempts where negligible or no change in the Casimir force was measured [1], or significant change was demonstrated only between different material systems [2].

In order to measure Casimir forces in PCMs, we prepared 1 μm thick amorphous AgInSbTe (AIST) thin films on 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 (see Fig. 11.1). For the crystalline sample the ellipsometry measurements were directly inverted to obtain the dielectric function $\varepsilon_\omega$ [13]. 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 optical properties of the substrate are only important in the IR range, where absorption of the film is weak. Thus it is justified to use tabulated data for the Al substrate.

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 is 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 [14]. As can be seen from Fig. 11.1, 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 [4, 5, 14]. The large change of the dielectric function upon crystallization suggests that a significant Casimir force contrast will be observed.

The measured dielectric response allows Casimir force calculations using the Lifshitz theory (Fig. 11.2), for which the force depends on the dielectric function at imaginary frequencies (inset Fig. 11.1). However, such forces are also affected by the surface roughness. The typical roughness of the samples was a few nm rms, but with a few isolated local peaks as evidenced by atomic force microscopy (AFM) analysis (lower inset in Fig.11.2). This small roughness is negligible for the Casimir force calculations in the measurement range but isolated peaks may slightly affect the force at the small distances.[17].

11.3 Measurements and comparison to theory
The Casimir force measurements, performed by G. Torricelli at the University of Leicester, are shown in Fig. 11.2. They were obtained using a dynamic AFM mode within an ultra high vacuum (UHV) Atomic Force Microscope (Omicron VT STM/AFM) [18, 19]. Forces were measured in the sphere-plate geometry between a Au coated (100 nm thick) 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 on different areas of both samples.

Figure 11.1: Absorptive part of the dielectric function for the crystalline (Δ) and amorphous (•) state of the AIST film obtained with ellipsometry as a function of frequency. The inset shows the same dielectric functions at imaginary frequencies $\zeta$, which is required for the Lifshitz theory.

Figure 11.2 Casimir force gradient measurement for the crystalline (Δ) and amorphous (•) phase. The theory curves are indicated with lines (—) 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 the topography measured by AFM of amorphous (left) and crystalline (right) films.
Figure 11.3 a): Determination of $V_0$ for the crystalline ($\Delta$) and amorphous (⋆) phase. The inset shows the force contribution due to varying $V_0$, divided by the Casimir force in percent. The latter indicates how much the remaining electrostatic interaction would contribute to the Casimir force if it was not subtracted. b) Comparison between the experimental electrostatic force gradient for $V_b=-50\text{mV}$ (⋆) and the theory taking into account the measured $V_0$ variations i.e. for $\Delta V=-50 \text{mV}$-$V_0(2)$ (−) and without i.e. $\Delta V=-50 \text{mV}$ (−).

The force measurement method and the experimental set-up are described in detail in [19]. Indeed, precise comparison of force measurements with theory is only possible if we 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 $V_0$ [19]. 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 [2, 18, 20] 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 $Z_0=42.8\pm0.5 \text{nm}$ for the amorphous, and $Z_0=42.9\pm0.4 \text{nm}$ 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 \( V_0 = 0 \) V), without the calibration being affected by variations in \( V_0 \). [19]. 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.11.2 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 qualitative agreement with the theoretical calculations. At short separations (< 55 nm) the increase in the difference is most likely to be attributed to the larger roughness of the crystalline state (lower inset Fig.11.2), leading to a larger force [17].

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 \( \sim 0.1 \) nm). In addition, it cannot be explained by the fact that the electrostatics have been performed using an approximate formula for capacitance gradient [19] which leads to an error of \( Z_0 \) of \( \sim 0.2 \) nm. Also, in order to check the force measurements we used a sample coated with low roughness Au (\( \sim 1 \) nm rms) and close agreement was found between the measured and theoretically predicted forces. Possible uncertainties in the optical properties of the PCM due to low and high frequency extrapolations, variation of the substrate properties or film thickness are 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 [21]. Indeed, the electrostatic force involves a larger interaction area on the plate than the Casimir force [21]. Larger areas contain more high peaks so that the averaged surface of the plate will be located higher than for smaller areas [21]. This is specific to the PCM roughness as the inset in Fig. 11.2 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 \( \sim 1-2 \) nm [21], 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 restored.
It is observed that there is a residual electrostatic force \( \sim V_0(Z)^2 \), where \( V_0 \) is the sphere-plate contact potential difference [19], which must be subtracted from the measured force. This is possible if \( V_0(Z) \) is known for all separations \( Z \) used for the force measurements. The variation of \( V_0(Z) \) can be extracted from the two electrostatic measurements (applied potentials \( V_b=\pm0.5V \)) by simple data manipulations (Fig. 11.3a) [19]. Variations for \( V_0 \) between 0-20 mV were observed for separations 40-150 nm without significant differences between amorphous and crystalline samples. As the inset in Fig. 11.3a indicates, subtraction of this residual electrostatic contribution corresponds to a correction of 6 % at \( Z=150 \) nm, and much less than 1% at \( Z=50 \) nm as compared to the Casimir force. Therefore, even avoiding this correction, the contrast of the force gradient between the two phases (inset Fig. 11.2) would remain practically unaffected. Finally, in order to fully confirm our force measurements, another electrostatic measurement was performed under identical conditions as before but with \( V_b=-50 \) mV (Fig. 11.3b). Again comparison of the force measurements with the theory, using the parameters extracted from the electrostatic calibration, shows very good agreement. Notably, as Fig. 11.3b shows, the agreement is better than that for the Casimir force measurements even though the force gradient for \( V_b=-50 \) mV is much smaller, confirming that the thermal drift is well compensated by the feedback loop.

11.4 Concluding remarks

As expected from the pronounced difference in the dielectric function of the amorphous and crystalline phases in phase change materials, a significant difference in the measured Casimir force between the PCM and Au was found for the two states. The measured force contrast is the largest reported to date for a switchable material [1]. 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. Indeed, the smaller the PCM cell the faster it can switch [15]. Switching times of a few nanoseconds already render phase change materials as very useful in electronic and optical memory applications [15]. Currently, there is a continuing effort to improve the number of switching cycles up to \( 10^{15} \) making for example PRAM (phase change random access memory) suitable to replace DRAM (Dynamic random access memory) [11]). The property portfolio of suitable dielectrical properties, fast switching, good scalability down to the nanometer regime [15], and strong Casimir force contrast deem PCMs to be promising (and potentially the only) candidates for a switchable Casimir force device.
References

The work described in this chapter has been published in:

