Measurement of the Casimir force between a spherical gold tip and Si(111)-(7×7) surfaces

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We have performed the measurement of Casimir force between a spherical Au tip and an atomically flat Si(111)-(7×7) surface at tip-sample distances ranging from 15 to 50 nm in an ultrahigh vacuum of 1.5×10^{-8} Pa by frequency-modulation atomic force microscopy. Atomically flat Si(111) surfaces provided by the ultrahigh-vacuum condition and a degassed Au tip reduce the contact potential difference that must be compensated. These experimental conditions led to the elucidation of the distance dependence of the Casimir force down to the distance of 15 nm. The observed distance dependence still follows a theory provided by Chen et al.[Phys. Rev. A **74**, 022103 (2006)] within these distances.

1. Introduction

When the separation between two uncharged metallic objects in micro-/nano-electromechanical systems (MEMS/NEMS) becomes a nanometer-scale, a quantum mechanical phenomenon called Casimir force prominently emerges as the force acting between the objects. The force predicted by Casimir is an attractive force induced by the exclusion of modes of electromagnetic fields in the region bounded by metallic plates.[1,2] In other words, the perturbation of zero-point vacuum fluctuations by conducting objects is the origin of the force. The force is sufficiently strong to be considered in a nanometer-scale separation embedded in nanoelectromechanical systems.[3,4] Novel ideas related to this force, such as quantum levitation[5–7] and conversion between electrical energy and quantum fluctuation energy in a vacuum,[8] have been proposed. To evaluate the strength of the Casimir forces experimentally, several measurements have been performed in the nanometer-scale over the past decade.[9–23] Experimental geometries using a metallic sphere and a metallic or dielectric plate have been employed in these attempts. The previous works on evaluating the Casimir force experimentally are summarized in Table I in accordance with their experimental conditions. The experimental conditions in this work are also listed in the table. Major differences between

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Table I. Measurement materials, distance range, and pressure in this work as well as in previous works.

Authors	Materials (plate-sphere)	Distance range (nm)	Pressure (Pa)	Year
Lamoreaux[9]	Au-Au	600 to 6000	1.3×10^{-2}	1997
Mohideen and Roy[10]	Al-Al	100 to 900	6.7	1998
Klimchitskyaya et al.[11]	Al-Al	80 to 910	6.7	1999
Harris et al.[12]	Au-Au	62 to 350	4.0	2000
Ederth[13]	Au-Au	20 to 100	Ambient	2000
Bressi et al.[14]	Cr-Cr (plate-plate)	500 to 3000	1.0×10^{-3}	2002
Decca et al.[15]	Cu-Au	200 to 2000	1.3×10^{-2}	2003
Chen et al.[16]	Si-Au	62 to 600	2.7×10^{-5}	2005
Chen et al.[17]	Si-Au	60 to 100	2.7×10^{-5}	2006
Chan et al.[18]	Si-Au	150 to 500	1.3×10^{-4}	2008
van Zwol et al.[19]	Au-Au	20 to 200	Ambient	2008
Banishev et al.[20]	Ni-Au	220 to 500	4.0×10^{-6}	2012
Chang et al.[21]	Au-Au	235 to 500	4.0×10^{-6}	2012
Laurent et al.[22]	Au-Au and Si-Au	100 to 400	4.0	2012
Banishev et al.[23]	SiO_2-Au	224 to 500	1.3×10^{-7}	2013
This work	Si-Au	15 to 50	1.5×10^{-8}	

the previous and our work are the range of observed distance and environment pressure. In the works summarized in Table I, the Casimir force was measured instead of the van der Waals force. The reason is that the Casimir force is thought to be the limiting case of the van der Waals force when the separation between the two bodies becomes large enough for retardation to be included.[16,17]

The Casimir force per unit area between two parallel plates having infinite permittivity and separated by the distance d_{pp} is expressed as $F_{pp} = -\pi^2 \hbar c / 240 {d_{pp}}^4$. It is strongly dependent on the distance d_{pp}; however, it is an experimentally difficult task to configure parallel plates separated by less than a nanometer-scale. Therefore, the forces between a large sphere and a flat plate were measured in the previous studies, except in the experiment reported in Ref. 14. The corresponding Casimir force was calculated as $F_{sp} = -\pi^3 R\hbar c / 360 {d_{sp}}^3$ on the basis of the proximity force theorem,[24] where R and d_{sp} are the radius of the sphere and the distance from the lowest surface of the sphere to the surface of the plate, respectively. The behavior of the Casimir force in a metal-semiconductor system is slightly different from that in a metal-metal system. Since the Casimir force is sensitive to the conductive properties of the semiconduc-

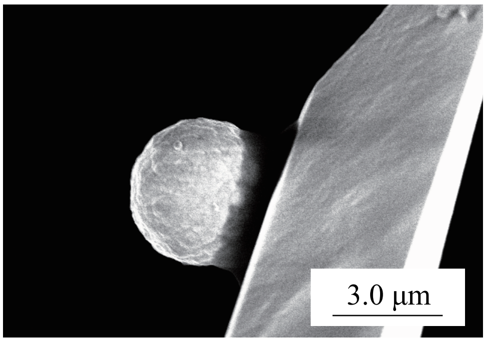

Fig. 1. (Color online) SEM image of the Au sphere mounted on the cantilever.

tors,[16,17,22,25,26] it should be possible to modulate the force in a metal-semiconductor system that is useful for NEMS applications. Therefore, we focus on the Casimir force between a metal and a semiconductor in this study. Spherical Au objects were widely used in the experiments listed in the table, and Chen et al. investigated the force using Si flat plates.[16,17] Since Au and Si have finite permittivity, the expression for the Casimir force between them should be modified. On the basis of the previous works on theoretical considerations where the finite conductivity was taken into account, Chen et al. expressed the Casimir force derived from the Lifshitz formula. We use this equation with the plasma frequency for Si (2.4×10^{13} rad/s) to evaluate our experimental results. The deviation from an ideal surface geometry or roughness should be considered to predict actual forces, as mentioned by Bordag et al.[27]

For experimental studies of the Casimir force, atomic force microscopy (AFM) has been widely used to evaluate the subtle force. To minimize capillary force due to water layers[28] on surfaces and improve the sensitivity of force-sensing cantilevers, AFM measurements have been performed in a vacuum, as seen in Table I. An ultrahigh vacuum (UHV) provides an atomically clean surface without contaminants that may cause residual patch charges and increase surface roughness. We have investigated the distance dependence of the force in a UHV with an atomically clean Si(111) surface at tip-sample distances shorter than those reported in the previous works. Furthermore, the distance dependence of force between a spherical Au tip and a Si(111)-(7×7) surface at distances ranging from 50 to 15 nm in a vacuum of 1.5×10^{-8} Pa is discussed.

2. Experimental procedure

A conventional frequency-modulation AFM (FM-AFM) system was used to investigate the force between a spherical tip and a flat surface in a constant-amplitude mode at

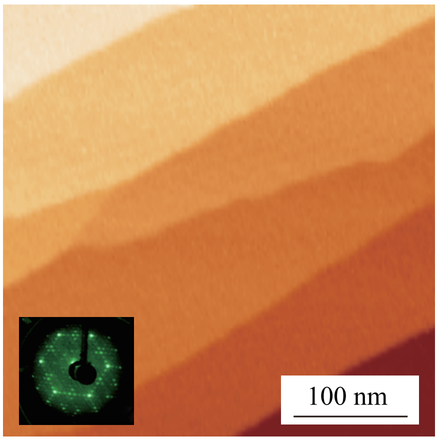

Fig. 2. (Color online) AFM image of Si(111) surface. The inset shows the low-energy electron diffraction pattern of the surface at 50 eV. It indicates a clean Si(111)-(7×7) surface.

room temperature in a UHV of 1.5×10^{-8} Pa. A Si(111) single-crystal substrate was used as the sample, and a commercially available cantilever with a spherical Au tip (sQube® colloidal probe) was employed. The nominal spring constant of the cantilever given on the data sheet was 4.2 N/m, and its mechanical resonant frequency was measured to be 84 kHz. The Q value of the cantilever measured under the UHV condition was 83000. This high Q value allows us to measure the weak force gradient necessary for evaluating the tip-sample interaction, such as the Casimir force.[29] The radius of the spherical Au tip, estimated by scanning electron microscope (SEM) observation, was 1.5 μm, as shown in Fig. 1.

Our measurement system (JEOL JAFM-4500XT) has a chamber for sample preparation, connected to an observation chamber equipped with the FM-AFM system. The preparation chamber has direct heating facilities for the Si(111) substrate to remove surface oxides and contaminants. The base pressure of both chambers was kept below 1.5×10^{-8} Pa. The Si(111) substrate was outgassed by heating at 833 K over two days and then flashed at 1453 K for 10 s. Then, the substrate was cooled slowly from 1173 to 873 K to obtain the 7×7 reconstructed and atomically flat surfaces. By using a conventional AFM cantilever with an atomically sharp tip (PPP-QNCHR-10) and from the low-energy electron diffraction pattern, the atomically cleaned Si(111)-(7×7) surface was observed, as shown in Fig. 2. After transferring the substrate to the observation chamber, the cantilever was heated to 423 K over two days to remove the surface adsorbates, including water molecules, in the preparation chamber.

In the FM-AFM experiments, the distance dependence of the frequency shift in the cantilever oscillation was measured to estimate the interacting force.[30,31] On the basis of the equation for conversion between frequency shift and force introduced by Sader et al.,[31] the force acting between the tip and the sample was calculated by integrating the frequency shift over the whole distance. In our practical calculation, the integral is bounded at the distance where the measured frequency shift becomes constant against distance variation.

In order to investigate the Casimir force in detail, other interaction forces should be excluded. In particular, the following dominant forces should be minimized. The first is capillary force,[28] and the second is the electrostatic force that arises from the contact potential difference (CPD) between the tip and the sample.[10,32] Since our measurements were carried out in a UHV environment after preparing the sample and the tip as described, the capillary force should be negligible. The bias voltage was applied between the tip and the sample to cancel the electrostatic force due to the CPD. To determine the value of the CPD, the amplitude shifts induced by the swept bias voltage were measured. If there are electrostatic patch potentials on the tip surfaces, the CPD may depend on the tip-sample distance and careful treatment would be needed to define its value.[33] To evaluate the influence of the patch potentials, the bias voltage was swept at five different distances. To avoid accidental crashing of the tip into the surface as a result of unstable feedback control in a small tip-sample distance range, small amplitude operation under the secondary resonant condition was employed. A higher mode has a higher spring constant and a lower mechanical quality factor, which are suitable for the small amplitude operation in dynamic force microscopy.[34] In this study, the amplitude shifts induced by the swept bias voltage were measured at the second resonant frequency (526 kHz).

3. Results and discussion

Figure 3 shows the amplitude shifts induced by the swept bias voltage on the Si(111) surface with the spherical Au tip at five different tip-sample distances. The CPD is defined as the bias voltage with which the amplitude is the maximum. The CPD should be carefully determined by assessing whether the CPD depends on the tip-sample distance; one should refer to the work performed by Inami and Sugimoto.[33] According to Inami and Sugimoto,[33] the CPD measurement based on the cantilever deflection produces an accurate CPD even if the CPD has a distance dependence. Since the distance dependence of the CPD is not notable in our CPD measurements based on the cantilever

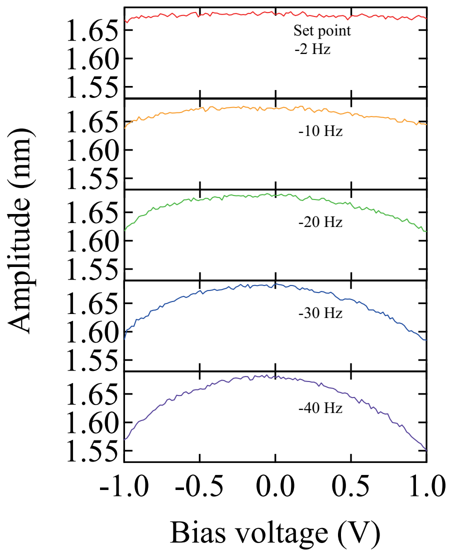

Fig. 3. (Color online) Amplitude shifts induced by the swept bias voltage at five set points in the oscillation of the second resonant frequency (526 kHz). The red, orange, green, blue, and purple curves are measured at the set points of -2, -10, -20, -30, and -40 Hz, respectively.

deflection, as shown in Fig. 3, the effect of the distance dependence of the CPD can be ignored in the tip-sample distance range in our case. All results of the measurements provide the maximum amplitude shift at approximately the same voltage of -50 mV. Therefore, the electrostatic force due to the CPD can be compensated by applying the bias voltage of -50 mV to the Si substrate.

To discuss the Casimir force acting between the tip and the sample, distance dependences of the amplitude and frequency shift of the oscillating cantilever were measured. Typical measurement results are shown in Figs. 4(a) and 4(b). The plots shown in the figures are the averages of 50 measurements at the same sample position. The horizontal axis shows the displacement of the sample estimated from the voltage applied to the the z-piezo positioner. We defined the origin of the distance as the inflection point of the frequency shift versus distance curves, where the amplitude is decreased by 0.6 nm from the amplitude of the constant state.

To convert the distance dependence of these parameters shown in Fig. 4 into the distance dependence of the force, the amplitude should be calibrated. Since an optical lever method is used to detect the oscillation of the cantilever in our experimental setup, the amplitude should be carefully calibrated. Our calibration procedure is as follows. After the first contact of the sample with the tip, the tip touches the surface of the sample during part of the oscillating period. Therefore, the Si(111) surface renders the slope of the intermittent contact equal to 1.[35] Considering this fact, the amplitude was calibrated using the slope of the intermittent contact to the horizontal axis. Note that the values on the horizontal axis are reasonable in the angstrom range, since it had been calibrated already from the atomic-resolution images.

In the previous works,[9–11] the distance dependence of the frequency shift (gradient of the force) and that of the calculated force were compared with the theoretical ones to evaluate the Casimir force. To do so, the origin of the tip-sample distance estimated from the experiments should have the same meaning as that in the theory. In this paper, we have defined the displacement of the sample as $d_{experimental}$ with its origin defined as the distance from the lowest surface of the tip to the surface of the sample. In the same way as in the other works, the tip-sample distance with the theoretical origin $d_{theoretical}$ should be obtained by adding an appropriate adjustment parameter d_0, that is, $d_{theoretical} = d_{experimental} + d_0$.

In other works,[9–11] d_0 was obtained by fitting the experimental force curve to the sum of the theoretical Casimir force and the electrostatic force caused by the residual CPD. In our case, d_0 was determined to be 12 nm by fitting the experimental force curve to the theoretical Casimir force, since the CPD was canceled by applying sample voltage. In the previous papers, the fitting parameters were given as 100,[9] and 120,[10] and 40 nm,[11] which means that the 12 nm used in our fitting is smaller than the previous ones.

This small value is due to the homogeneity of the Si(111) surface. The conventional FM-AFM observation revealed that the roughness of the Si surfaces is about 0.7 nm. Since the clean Si(111) surface is rather flat compared with the roughness of the Au sphere, the nonzero d_0 value could have originated from the nonuniformity of the Au sphere on the cantilever. The contribution of the residual CPD to the determination of d_0 can be ignored because CPD did not show clear distance dependences, as discussed above. Because the average deviation of the Au sphere from the ideal one is roughly estimated to be less than 20 nm by SEM observation, d_0 is thought to be the distance at the time of contact owing to the sphere roughness.[19] Therefore, it is reasonable that d_0 is of the same order as the average deviation.

In Fig. 5, the distance dependence of the force between the spherical Au tip and the Si flat surface is shown. Here, the adjustment parameter d_0 was taken into account, as described above. The solid curve shows the distance dependence of the theoretically calculated Casimir force by using the equations described in Ref. 17 with our experi-

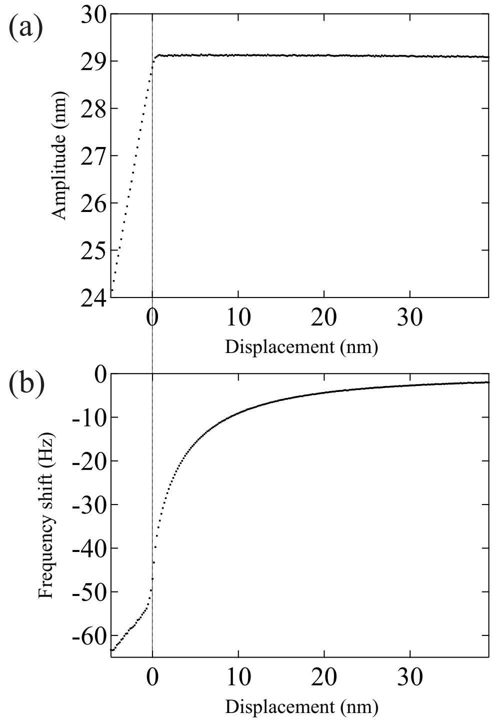

Fig. 4. (a) Distance dependence of the vibration amplitude of the cantilever. (b) Distance dependence of the frequency shift of the cantilever.

mental conditions. In the distance range from 15 to 50 nm, the observed forces fit to theoretical ones.

Chen et al.[17] showed that their experimentally estimated forces between Au and Si materials are well predicted by theoretical calculation of the Casimir force within the distance range from 60 to 100 nm with a roughness correction. Considering that we used the theoretical calculation expressed in the paper, our results shown in Fig. 5 indicate that the observed force was caused predominantly by the Casimir force, because the observed force well fits the calculated force within the distance range from 15 to 50 nm. Since the roughness of the Au sphere is comparable to the shortest distance of the measurement range shown in Fig. 5, the roughness correction proposed in Ref. 17 should not be applied in our case. Since the atomically clean and flat Si(111) surface of the sample has a small roughness that can be ignored in our analysis, only the surface roughness of the tip should be considered in determining d_0.

At a distance shorter than 15 nm, the observed forces deviate from the theoretically calculated forces (see the inset in Fig. 5). To understand the reason for this deviation, a theory used to treat imperfections of the spherical Au tip surface or roughness of the

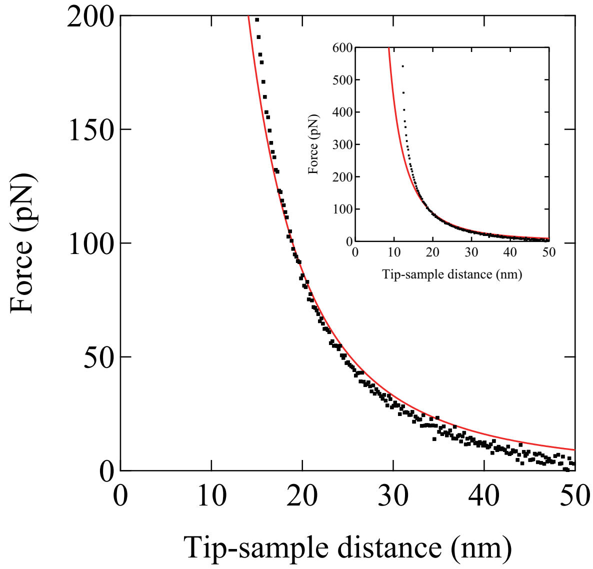

Fig. 5. (Color online) Force versus tip-sample distance. The square dots and the solid line show the measured curve and the theoretically calculated Casimir force, respectively.

tip in this distance range should be developed and the experiment using a smoothly rounded spherical metallic tip should be performed. In our measurement, the small protrusion at the tip apex could act as an effective tip at distances less than 15 nm and cause the deviation.

4. Conclusions

We measured the interaction force dominantly caused by the Casimir force between a spherical Au tip and an atomically flat Si(111)-(7×7) surface at tip-sample distances ranging from 15 to 50 nm in an ultrahigh vacuum of 1.5×10^{-8} Pa by FM-AFM. In the previous studies on the Casimir force between a metal and a semiconductor, the experiment was performed in a vacuum of 2.7×10^{-5} Pa at distances down to about 60 nm.[17] The ultrahigh-vacuum condition provides atomically clean and flat Si(111) surfaces. In addition, a careful degassing treatment of the Au spherical tip reduces the residual CPD that must be compensated. These experimental conditions enable us to reveal the distance dependence of the force predominantly caused by the Casimir force down to the distance of 15 nm. The theory described in Ref. 17 enables good understanding of the distance dependence; however, more detailed discussion, including that on the roughness of the tip surface, which is comparable to the tip-sample distance, is needed to explain the deviation of the experimental results from the theoretically estimated values at the smaller distances.

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