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Electrically switchable Casimir forces using transparent conductive oxides

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Abstract

Casimir forces between charge-neutral bodies originate from quantum vacuum fluctuations of electromagnetic fields, which exhibit a critical dependence on material's electromagnetic properties. Over the years, *in-situ* modulation of material's optical properties has been enabled through various means and has been widely exploited in a plethora of applications such as electro-optical modulation, transient color generation, bio- or chemical sensing, etc. Yet Casimir force modulation has been hindered by difficulty in achieving high modulation signals due to the broadband nature of the Casimir interaction. Here we propose and investigate two configurations that allow for *in-situ* modulation of Casimir forces through electrical gating of a metal-insulator-semiconductor (MIS) junction comprised of transparent conductive oxide (TCO) materials. By switching the gate voltage on and off, a force modulation of > 400 pN is predicted due to substantive charge carrier accumulation in the TCO layer, which can be easily measured using state-of-the-art force measurement techniques in an atomic force microscope (AFM). We further examine the influence of the oxide layer thickness on the force modulation, suggesting the importance of the fine control of the oxide layer deposition. Our work provides a promising pathway for modulating the Casimir effect *in-situ* with experimentally measurable force contrast.

Introduction

Quantum vacuum fluctuations of electromagnetic fields are a fascinating quantum-mechanical effect, manifested by a multitude of celebrated physical phenomena such as Lamb shift, spontaneous emission, the anomalous magnetic moment of electron and surface wetting [1]. Amongst them is the Casimir effect, named after H. B. G. Casimir who, in 1948, predicted an attractive force between two perfectly conducting parallel plates that scales with the plate-plate separation d as $\propto d^{-4}$ [2]. This force, manifested as a macroscopic quantum effect, was first calculated by considering the perfectly reflective boundary condition of ideal metal plates imposed on the quantum vacuum fields, which alters the spatial distribution of the zero-point energy density compared with free space by quantum field theory. Since the discovery, the Casimir effect has been of fundamental research interest on its own as well as through its connection to other fields in fundamental physics (e.g., exploration of gravity at the microscale, search for extra forces, and testing of the prediction of new physics beyond the Standard Model [3-7]). In the meantime, it has also brought about significant implications in nanotechnology, particularly in micro/nanoelectromechanical systems (MEMS/NEMS) where devices are engineered with movable parts on the micro/nanoscale such that those quantum effects become significant [8-11].

45 As a direct manifestation of the boundary condition dependence of the quantum fluctuations,
46 if the perfectly reflective boundary of the interacting bodies is relaxed to a finite conductivity, the
47 vacuum fluctuation interaction between two bodies can be described approximately by impedance
48 boundary conditions with finite penetration depth. Alternatively, the Casimir effect can be
49 interpreted as resulting from the coherent oscillations of dipole moments of a large number of atoms
50 in the bodies, which renders the material influence on the force in a nonintuitive manner. Most
51 generally, the calculation of this effect should rigorously consider the interaction energy with the
52 frequency-dependent dielectric function of the involved materials. Consequently, the magnitude
53 and/or the sign of the Casimir force can be dramatically altered if the interacting materials (and the
54 intervening medium) are appropriately chosen [12-21]. For instance, it has been demonstrated that
55 the Casimir force between an Au-coated sphere and a transparent conductive oxide (TCO) film is
56 nearly half of the value found between two noble metal films [14,22]. On the other hand, the force
57 between an Au sphere and a silica film immersed in certain liquid solutions (e.g., Bromobenzene)
58 were measured to be repulsive [16].

59 The material dependence of the Casimir force has also provoked the pursuit of direct force
60 modulation by modifying the optical properties of the materials. Modulation of the Casimir force is
61 of potentially profound technological significance in MEMS/NEMS. For example, unwanted
62 stiction or adhesion between movable parts can occur due to Casimir interactions as MEMS/NEMS
63 devices continue to miniaturize [8,9,23]. Reduction in force magnitude is paramount to mitigate
64 these issues. On the other hand, the Casimir effect can also be exploited as an external force to
65 actuate the micro- and nano-devices with quantum fluctuations where increased force magnitude
66 may be desired [24-26]. Appropriate doping in semiconductors can readily modify the charge carrier
67 density, giving rise to notable alteration of their optical properties and the resulting forces [27-29].
68 In addition, marked force contrast has been demonstrated or predicted for configurations based on
69 phase-change materials in their different states [30-35].

70 However, the abovementioned techniques usually require non-trivial thermal treatment and the
71 force modulation is not *in-situ*. *In-situ* force modulation helps to control the actuation dynamics
72 through dynamic switching between high-low force states, which is indispensable for many
73 MEMS/NEMS devices (e.g., switches, oscillators, parametric amplifiers, nanometric position/force
74 sensors, etc.) to properly operate or to combat unwanted stiction between adjacent components [36-
75 38]. To date, *in-situ* Casimir force modulation has mostly been carried out through the drive of
76 mechanical motion of one of the bodies. There have been few attempts at *in-situ* Casimir force
77 modulation in response to external stimuli due to experimental difficulties that arise when
78 modulating the optical properties of materials in Casimir measurement configurations. Chen *et al.*,
79 for example, have achieved optical modulation of the Casimir force with up to a few pico-Newtons
80 variation between an Au-coated sphere and a single-crystalline Si membrane through the excitation
81 of charge carriers in the semiconductor using a pulsed Ar laser [39,40]. However, laser-induced
82 Casimir force modulation undergoes undesired artifacts such as heating and exerted optical forces,
83 which can further complicate the experimental consideration. Alternately, using phase-change
84 materials for *in-situ* operation is anticipated to face substantial challenges due to protective layers
85 and volume compression upon phase transition [31,37].

86 From the perspective of charge carrier density modulation, electrical biasing/gating is a high-
87 speed modulation technique which is generally easier to operate, less power-consuming and less
88 prone to the above-mentioned artifacts compared to many other techniques. In particular, metal-

89 insulator-semiconductor (MIS) junctions comprising TCOs such as ITO (*i.e.* indium tin oxide) have
90 been widely employed in high-speed electro-optical modulators where the optical responses of the
91 devices can be adapted *on-demand* by tuning the gate voltage, as ITO exhibits gate-controllable
92 optical properties through charge carrier accumulation or depletion at ultrafast speed [41-47]. From
93 the perspective of the intrinsically broadband nature of Casimir effect, profound modification of the
94 optical property from the IR up to the UV with the change of carrier density would also render ITO
95 a great candidate material for modulating the force [14,48]. However, studies on gating-enabled
96 Casimir force modulation are sparse. One recent theoretical work reported the Casimir interaction
97 between a gold platelet and a multilayer stack consisting of a MIS junction made of ITO-Teflon-
98 gold immersed in a liquid environment. It was predicted that the platelet can switch between a
99 “trapped” state and a “released” state by varying the charge carrier density in the ITO layer [49].
100 Nonetheless, to the best of our knowledge, gate-switchable Casimir forces in an experimentally
101 amenable configuration with pragmatic material and structural parameters and sufficiently
102 measurable force contrast between the “on” and “off” state are still missing.

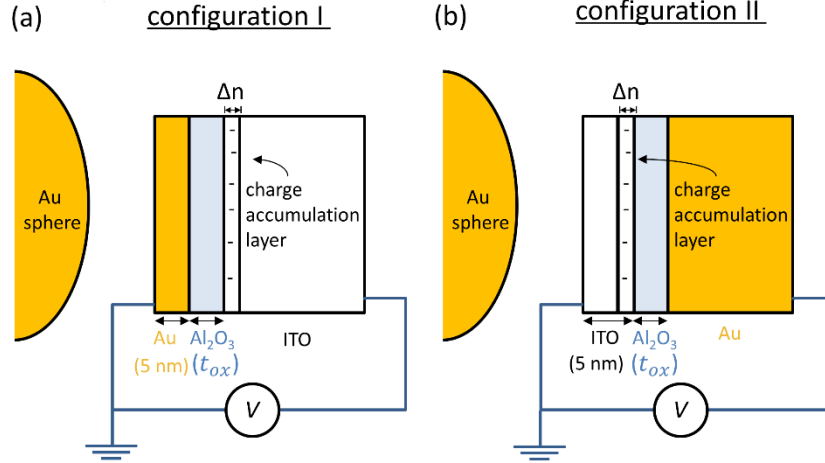
103 In this work, we propose two configurations to realize gate-switchable Casimir forces which
104 can be directly deployed in well-established experimental setups. For both configurations the
105 Casimir interaction would be measured between an optically thick Au film-coated sphere, which
106 could be attached to an atomic force microscope (AFM) cantilever for force detection, and a gate-
107 controlled MIS junction consisting of Au-Al₂O₃-ITO planar films with an applied gate voltage. Two
108 potential configurations are considered (Fig. 1). In configuration I, the ITO film is optically thick
109 and coated by ultrathin layers of Al₂O₃ and Au. Configuration II is inverted, with a thick film of Au
110 coated with ultrathin layers of Al₂O₃ and ITO. With a reasonable gate voltage range (0-6 V), the
111 charge carrier density in the ITO accumulation layer can increase by more than an order of
112 magnitude from 10¹⁹ cm⁻³ to (4-6)×10²⁰ cm⁻³. At short separations (10-50 nm) between the sphere
113 and the MIS stack, the force modulation magnitude is found to reach up to ~15 pN for configuration
114 I and to up to > 400 pN for configuration II, both of which far exceed the measurement sensitivity
115 of the state-of-the-art force measurement techniques using an AFM. Further, we find the thickness
116 of the ultrathin oxide layer between the two electrodes plays a significant role in determining the
117 modulation strength, whose value is enhanced by up to 1.7 times when the thickness is reduced from
118 3 nm to 2 nm. Our results demonstrate the intriguing prospect of achieving high-speed switchable
119 Casimir forces *in-situ* through electrical gating and provide a rational design for future
120 experimentation.

121

122 **Results and discussions**

123 Figure 1 illustrates the two configurations mentioned above. The radius of the Au-coated
124 sphere is set to be 100 μm, a common value reported in literature [14,27,39,48,50-54] for Casimir
125 force measurements using an AFM. The spherical geometry avoids the alignment problem for two
126 large parallel plates and has been a well-established force measurement configuration in AFM. The
127 Al₂O₃ layer thickness t_{ox} in the MIS junction is set to be 3 nm, which can be precisely controlled
128 using atomic layer deposition (ALD) technique [55-57]. The top coating layer (Au for configuration
129 I and ITO for configuration II) is set to be thin (5 nm) to warrant a sufficiently large modification to
130 the force while ensuring reasonably good conductivity of the film [43,46,58-60]. In both
131 configurations, the ITO serves as both the active layer for charge carrier density modulation and the
132 electrode for applying gate voltage. Besides, ITO conducts sufficiently well to eliminate surface

133 trap charges which would otherwise obscure the measurement of the Casimir force [14]. Note that
 134 the top layer (Au in configuration I and ITO in configuration II) is grounded and therefore a negative
 135 bias is applied in configuration I whereas a positive bias is applied in configuration II to the substrate
 136 to form the charge accumulation layer at the interface between ITO and the oxide, which is typically
 137 1-3 nm thick [42-46,61,62].
 138



139
 140 **Figure 1.** Two proposed configurations for actively switchable Casimir forces. A gold sphere of radius $R=100 \mu\text{m}$
 141 is brought close to a metal-insulator-semiconductor (MIS) junction consisting of an Au layer and an ITO layer
 142 sandwiching an Al_2O_3 ultrathin film. When the junction is gated, the charge carrier density at the interface between
 143 the oxide and ITO is significantly increased, forming an ultrathin accumulation layer with modified optical
 144 properties compared to the otherwise as-deposited ITO film due to the charge accumulation. The Casimir force
 145 between the Au sphere and the MIS junction is thus modified. The orientation of the MIS junction is different for
 146 the two configurations, as (a) the Au film faces the Au sphere and (b) the ITO layer faces the Au sphere.
 147

148 To quantify the charge accumulation effect at the ITO-oxide interface, we utilize a simple
 149 capacitance model across an MIS junction which assumes a uniform carrier density in the ultrathin
 150 accumulation layer, as widely adopted in literature [42,47,60-62]. The average thickness t_{acc} of
 151 the accumulation layer due to carrier injection in a standard MIS junction is given by [63]: $t_{acc} =$
 152 $\frac{\pi}{\sqrt{2}} \sqrt{\frac{k_B T \epsilon_0 \epsilon_S}{N_0 q^2}}$, where k_B is the Boltzmann constant, $T = 300 \text{ K}$ is the room temperature, ϵ_0 is the
 153 free-space permittivity, $\epsilon_S = 9.3$ is the relative static permittivity of ITO [42,44,46], q is the
 154 electron charge, and N_0 is the initial carrier density in the ITO layer. In practice, the carrier density
 155 in ITO as-deposited is dependent upon the deposition processes and annealing conditions [64,65],
 156 thus can vary by as large a range as 10^{19} - 10^{21} cm^{-3} . We set the ITO initial carrier density as 1×10^{19}
 157 cm^{-3} for our computation, the same as reported in literature [46,47,61], which yields $t_{acc} =$
 158 2.56 nm . When a gate voltage V_g is applied across the junction, the carrier density in the
 159 accumulation layer can be written as:

$$160 \quad N_{acc} = N_0 + \frac{\epsilon_0 \epsilon_{ox} V_g}{q t_{ox} t_{acc}} \quad (1)$$

161 where $\epsilon_{ox} = 9$ denotes the relative static permittivity of Al_2O_3 [44,60,66]. Here we restrict $V_g <$
 162 6 V to avoid electrical breakdown of the oxide [56,57,67], which increases the carrier density in
 163 the accumulation layer to about $4 \times 10^{20} \text{ cm}^{-3}$, more than an order of magnitude larger than the initial

164 value. Such a profound modulation of carrier density *via* gating in ITO-based MIS junctions has
 165 also been reported by a number of experimental works [41,46,47,61,68].

166 The Casimir force between a sphere with radius R and a planar structure at a separation d is
 167 given by the Lifshitz formula using proximity force approximation (PFA) provided $R \gg d$ [8,69]:

$$168 \quad F(d) = k_B T R \sum_{m=0}^{\infty} \int_0^{\infty} k [\ln(1 - r_1^{TE} r_2^{TE} \exp(-2k_{\perp} d)) + \ln(1 - r_1^{TM} r_2^{TM} \exp(-2k_{\perp} d))] dk \quad (2)$$

169 where k is the lateral wavenumber, $k_{\perp} = \sqrt{k^2 + \frac{\xi_m^2}{c^2}}$ is the vertical wavenumber in the intervening

170 medium (air), $\xi_m = \frac{2\pi k_B T}{\hbar} m$ denotes the Matsubara frequencies, the prime sign on the summation

171 means the zero-frequency term is multiplied by half, and r_i^{σ} ($i = 1, 2$ and $\sigma = TE, TM$) represent

172 the reflection coefficients at the interface between air and medium i (note: the Au sphere is medium

173 1 and the MIS stack is medium 2 for our configuration) for imaginary frequency ξ_m and lateral

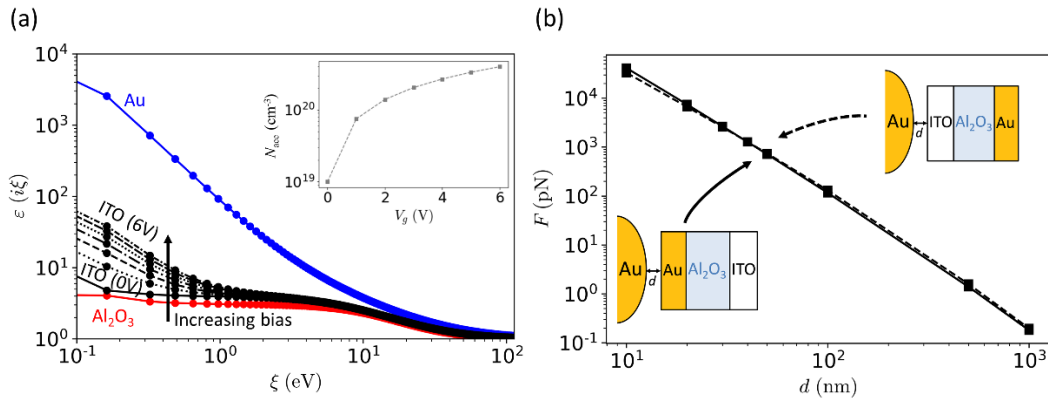
174 wavenumber k under TE and TM polarizations. The reflection coefficients off the surface of the

175 stack are computed using the transfer matrix method (TMM) [49]. Because the reflection depends

176 naturally on the material's broadband dispersion (dielectric function) and the object's geometry and

177 size, so does the resulting force.

178



179

180 **Figure 2.** (a) Dielectric functions for different materials with respect to the Matsubara frequencies at room
 181 temperature. The dielectric function of the ITO accumulation layer monotonically increases with applied gate
 182 voltage (oxide thickness is 3 nm in the MIS junction). Inset shows the carrier density increase in the accumulation
 183 layer in ITO with applied gate voltage. (b) Casimir force between the sphere and the MIS stack under zero gate
 184 voltage as a function of separation. The solid and dotted line represent the force for configuration I and II,
 185 respectively.

186

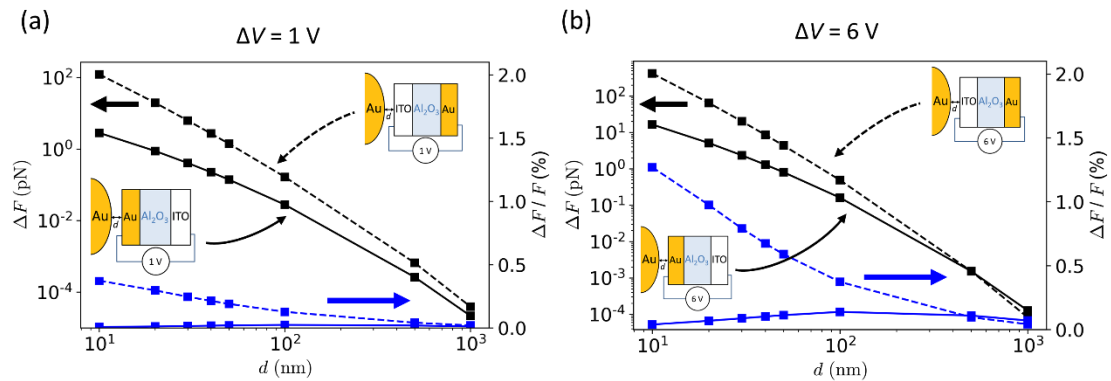
187 We apply dielectric function data/models for the materials using the most often utilized data
 188 for Casimir force calculations. The optical data for Au is obtained from Palik's handbook, extended
 189 to lower energies using a Drude model with parameters $\omega_p = 9$ eV and $\gamma_p = 0.035$ eV
 190 [13,14,27,54,70,71]. The dielectric function for Al_2O_3 is modeled using a dual-oscillator Lorentz
 191 model [12,72]. For ITO, we apply the dielectric function constructed by the sum of Drude and Tauc-
 192 Lorentz models using the parameters found in the literature [14,54]. In the Drude term, the plasma

193 frequency is directly related to the charge carrier density by $\omega_p = \sqrt{Nq^2/\epsilon_0 m^*}$, where N is the

194 charge carrier density and m^* is the charge carrier effective mass. Figure 2a shows the dielectric

195 functions of the abovementioned materials. As expected, the permittivity values with respect to
 196 Matsubara frequencies for the accumulation layer in ITO lie between those for Au and Al₂O₃ and
 197 monotonically increase with applied gate voltage as a result of augmented carrier density, which
 198 renders the interface more “metallic”. The calculated Casimir forces for both configurations under
 199 zero gate voltage are shown in Fig. 2(b). We note that they exhibit commensurate force magnitudes
 200 in this separation range.

201 When a gate voltage is applied across the junction, the force magnitude is modified due to the
 202 change of charge carrier density in the accumulation layer (Fig. 3), which ultimately alters the
 203 overall reflection at the top surface of the stack. The force modulation ΔF (compared with zero
 204 gate voltage) is over an order of magnitude larger for configuration II compared to configuration I.
 205 Further, we find that the force modulation reaches > 400 pN when the separation is reduced to 10
 206 nm with an applied bias of 6 V. Contrastingly, the force modulation is much less than 1 pN with
 207 separations greater than 50 nm. Note that the positive values for ΔF means the force becomes more
 208 attractive when gate voltage is turned on, in agreement with the intuition that the stack becomes
 209 more metallic as a result of charge carrier injection. We also note that while the absolute force
 210 modulation ΔF always decreases monotonically with increasing separation, the relative
 211 modulation $\Delta F/F$ behaves differently for the two configurations: In configuration I, $\Delta F/F$
 212 reaches optimal values at an intermediate separation (on the order of 100 nm), whereas $\Delta F/F$
 213 monotonically rises with reduced separation in configuration II, reaching the value of $\sim 1.3\%$, larger
 214 than the highest reported 1% for *in-situ* force modulation to the best of our knowledge [39].
 215



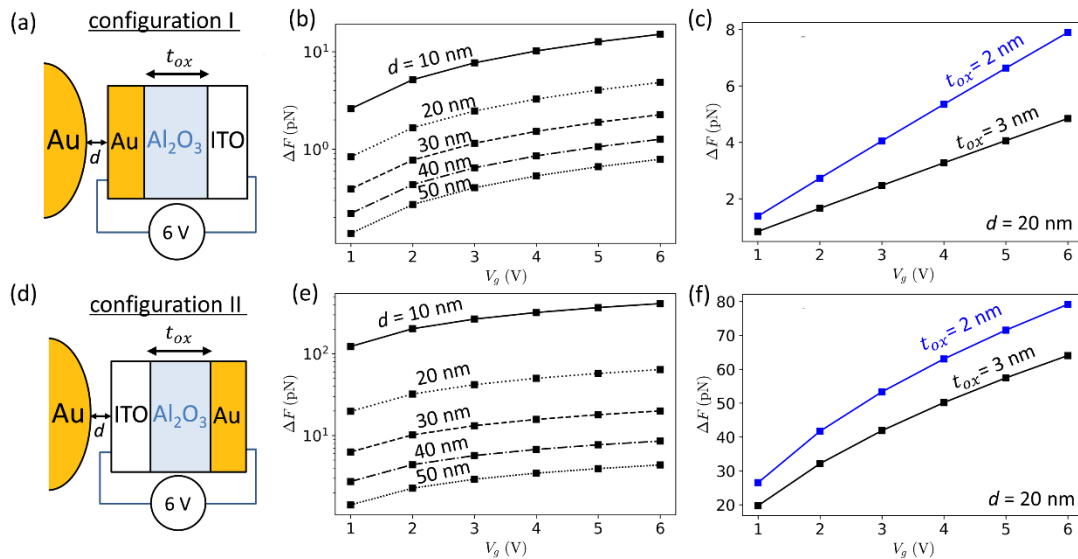
216
 217 **Figure 3.** Force modulation as a function of separation under the gate voltage of (a) 1 V and (b) 6 V. The force
 218 change in configuration II is on average more than one order of magnitude larger than in configuration I. The solid
 219 and dotted lines represent the force modulation for configuration I and II, respectively. Black (for left vertical axes)
 220 and blue (for right vertical axes) lines represent the absolute and relative force modulation, respectively.

221
 222 At a fixed separation, the force modulation varies as a function of both the applied voltage bias
 223 and insulating layer (Al₂O₃) thickness (Fig. 4). We find similar behavior for configurations I and II
 224 (Fig. 4a and Fig. 4d, respectively), with more pronounced variations for configuration II. For both
 225 configurations, ΔF monotonically increases with increasing gate voltage due to the enhanced
 226 reflection of the structure with increased carrier density. Figure 4c shows how the force modulation
 227 is controlled by the gate voltage at a separation of 20 nm with two different oxide thicknesses. The
 228 reduction of the thickness from 3 nm to 2 nm enhances the modulation magnitude by more than
 229 60%, resulting in $\Delta F \sim 8$ pN for a 6 V gate voltage. The strong dependence of the force change on

230 the oxide thickness is attributed to the change of carrier accumulation at the ITO-oxide interface.
 231 With a 2-nm thick oxide layer, the carrier density reaches $5.93 \times 10^{20} \text{ cm}^{-3}$ with a 6 V gate voltage,
 232 about 1.5 times that for a 3-nm oxide layer. One caveat of utilizing a thinner oxide layer is that the
 233 maximum gate voltage to be applied is further constrained by the breakdown field strength of the
 234 oxide. Fortunately, precise control of the oxide layer at the level of sub-nanometer scale is made
 235 possible by advanced deposition techniques such as ALD [67].

236 Compared with configuration I, the modulation for configuration II (Fig. 4(d)-(f)) is on average
 237 one order of magnitude stronger in the separation range we considered, which allows for a
 238 measurable force modulation even with just 1 V gate voltage switched on and off. This behavior can
 239 be ascribed to the closer distance between the charge accumulation layer and the Au sphere, leading
 240 to a much greater reflection change at the top surface of the stack. Because the force modulation
 241 magnitude is significantly greater for this configuration, we anticipate that configuration II will be
 242 much easier to embody in experiment. Likewise, reduction of the oxide thickness to 2 nm increases
 243 ΔF by 1.2-1.3 times, as shown in Figure 4f. One interesting visual distinction between the two
 244 configurations is how ΔF scales with the gate voltage V_g (and the resulted variation of the carrier
 245 density N_{acc}). In configuration I, ΔF increases almost linearly with V_g . Contrastingly, the
 246 variation of ΔF with V_g is more nonlinear in configuration II. Nonetheless, the visually perceived
 247 linearity for configuration I is merely the result of very small force modulation values. This behavior
 248 is another manifestation of the highly complex nature of the relation between material's local optical
 249 properties and the force.

250



251

252 **Figure 4.** Modulation of the Casimir force with applied gate voltage and oxide layer thickness. (a) Schematic of
 253 configuration I showing (b) the force change at different separations with an oxide thickness of 3 nm in the MIS
 254 junction. (c) Force change at a fixed separation of 20 nm, with two different oxide thicknesses (2 nm and 3 nm,
 255 respectively). (d,e,f) Same as (a-c) but for configuration II.

256

257 From an experimental point of view, state-of-the-art AFM techniques with a sphere-planar
 258 configuration feature a force measurement sensitivity of 1-5 pN [73-76] to as small as a few fN
 259 [28,77]. This indicates that to obtain a measurable force modulation, the less sensitive measurement
 260 techniques would require a separation of less than 30 nm while the gate voltage is switched between

261 0 and 6 V. To reach these small separations, which have been achieved in other experiments
262 [12,53,76,78,79], the jump-to-contact (JTC) distance and the surface roughness should be reduced.
263 This reduction can be obtained by increasing the cantilever stiffness and by reducing the sphere size.
264 While increasing the cantilever stiffness can also reduce the sensitivity, there are ways to counteract
265 this reduction. Because the force modulation is generated by switching the gate voltage on and off,
266 it can, in principle, be directly measured with a better sensitivity using a lock-in amplifier by
267 referencing the voltage on-off control signal at a particular modulation frequency (~100-1000 Hz)
268 for phase-locking. In fact, Chen *et al.* employed a similar technique to measure the laser-induced
269 force modulation, reducing the measurement noise to the level of 0.1-0.5 pN [39,40]. As a final
270 comment about the potential experimental realization of electrical modulation of the Casimir force,
271 care must be taken to ensure that no residual electrostatic forces obscure the measurement.
272 Electrostatic force cancelation is typically performed by applying a counter-bias between the sphere
273 and a grounded plate. For the configurations that we propose, the plate closest to the sphere could
274 be grounded and the junction bias can be applied via the back electrode relative to this ground. In
275 that way, two counter-biases can be applied: one to the sphere and one to the backside of the junction.
276 It was also found in a previous experiment with laser illumination that a variation of the charge
277 density can result in a modification to the residual electrostatic potential, which can be nullified
278 during the experimental procedure [39]. We note that even after compensation, there can still exist
279 a voltage error of the order 0.4 ~1.5 mV [22,48,54]. Assuming an error of 1 mV, the residual
280 electrostatic force at varying sphere-plate separation is calculated to be 0.1-0.28 pN at separations
281 below 30 nm. Consequently, the measurement of modulated force would not be obscured by the
282 uncertainty due to the electrostatic force provided proper voltage compensation is applied.

283 284 **Conclusion**

285 In summary, we theoretically investigated two configurations for potential implementation of
286 gate-switchable Casimir forces, both of which are composed of a gate-controlled MIS junction of
287 Au-Al₂O₃-ITO planar films, with different orientations towards a gold-coated sphere attached to an
288 AFM cantilever. The charge carrier density in the ITO accumulation layer formed at the interface
289 between ITO and the oxide layer can be tuned substantially from 10¹⁹ cm⁻³ to (4-6)×10²⁰ cm⁻³ *via*
290 gating. As a result, a force modulation magnitude reaches up to > 400 pN with a gate voltage of 6
291 V, far exceeding the measurement sensitivity with the state-of-the-art AFM force-measurement
292 techniques. Furthermore, a reduction of the oxide layer thickness from 3 nm to 2 nm can increment
293 the force modulation magnitude by up to 70%, which indicates the precise control of the oxide layer
294 thickness *via* advanced deposition techniques such as ALD is paramount for force modulation. Our
295 results show the great promise of utilizing TCO materials to realize switchable Casimir forces with
296 a pronounced force contrast, which may create new opportunities for *in-situ* control and modulation
297 of movable parts in nanomechanical devices and systems.

298 299 **Acknowledgements**

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