

Electric double layer effect in an extreme near-field heat transfer between metal surfacesA. I. Volokitin ^{*}*Department of Physics, Samara State Technical University, 443100 Samara, Russia*

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Calculations of heat transfer between two plates of gold in an extreme near field is performed taking into account the existence of the electrical double layer on metal surfaces. For $d < 3$ nm the double layer contribution to the heat flux exceeds the predictions of the conventional theory of the heat transfer by several orders of magnitudes. The predicted effect is not related with bias voltage but occurs due to intrinsic properties of the electrical double layer. The results obtained can be used for the heat management at the nanoscale by intentionally changing the parameters of surface dipoles with the help of engineering.

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All bodies are surrounded by a fluctuating electromagnetic field due to quantum and thermal fluctuations. This electromagnetic field is responsible for the fluctuation-induced electromagnetic phenomena (FIEP), such as the Casimir force, that play an extremely important role in nanotechnology because it is one of the dominant forces at the nanoscale. At present a great deal of attention is attracted for studies of FIEP at dynamic and thermal nonequilibrium conditions. Under these conditions, a modification of the Casimir force appears and new effects arise such as the radiative heat transfer and Casimir friction, and it becomes possible to control FIEP, which is extremely important for the development of micro- and nanoelectromechanical devices. FIEP are usually described by a fluctuation electrodynamics developed by Rytov [1–3]. In the framework of this theory it was theoretically predicted [4–8] and experimentally confirmed [9–14] that the radiative heat flux between two bodies with different temperatures in the near field (when the distance between the bodies $d < \lambda_T = c\hbar/k_B T$: At room temperature $\lambda_T \sim 10$ μm) can be by many orders of magnitude larger than the limit, which is established by Planck’s law for blackbody radiation. With the development of new experimental techniques over the past decade, super-Planckian heat transfer has been observed for vacuum gaps between bodies in the interval from hundreds of nanometers to several Ångströms [11–14]. Generally, the results of these measurements turned out to be in good agreement with the predictions based on the fluctuation electrodynamics for a wide range of materials and geometries. However, there still remain significant unresolved problems in understanding heat transfer between bodies in an extreme near field (gap size < 10 nm) [12,13]. In Refs. [12,13] the heat flux between a gold coated near-field scanning thermal microscope tip and a planar gold sample in an extreme near field at nanometer distances of 0.2–7 nm was studied. It was found that the experimental results can not be explained by the conventional theory of the radiative heat transfer based on Rytov theory. In particular, the heat transfer observed in

Ref. [12] for the separations from 1 to 10 nm is orders of magnitude larger than the predictions of conventional Rytov theory and its distance dependence is not well understood. These discrepancies stimulate the search of the alternative channels of the heat transfer that can be activated in the extreme near field. One of the obvious channels is related with “phonon tunneling” which stimulated active research of the phonon heat transfer in this region [15–28]. In Refs. [15,16] it was shown that the heat flux in the extreme near field can be strongly enhanced in the presence of the potential difference between bodies. This enhancement is due to the electric field effect related to the fluctuating dipole moment induced on the surface by the potential difference. In this Letter another mechanism, related to the fluctuating dipole moment of the electric double layer on a metal surface, is considered. In contrast to the electric field effect, which is short range and operates for the separations of order ~ 1 nm, the double layer effect is long range and operates for the separations $< 10^2$ nm.

The electric double layer on the metal surface is associated with a redistribution of the electron density, as a result of which the surface atomic layer 1 is charged negatively, and the near-surface layer 2 is positively charged (see Fig. 1). The generic mechanism for such redistribution for metal surfaces is the “spill out” of electron into vacuum. Deep in the metal the ion charges are compensating by negative electron density, but as the lattice abruptly terminates at the surface, electrons tunnel out the solid over some small distance (Ångström), creating a negative sheet of charges in the surface atomic plane and leaving a positive sheet of uncompensated metal ions in the subsurface atomic planes [29,30]. The electric double layer can be approximately considered as formed by two oppositely charged planes with a surface density $\pm\sigma_d$ (see Fig. 1). This double layer of charges creates a potential step $\Delta\varphi$ that increases the electron potential just outside the surface, effectively raising work function [30]

$$A = \Delta\varphi - \bar{\mu}, \quad (1)$$

where $\bar{\mu} = \mu - \bar{\varphi}$ is bulk chemical potential measured from the level of mean electrostatic potential.

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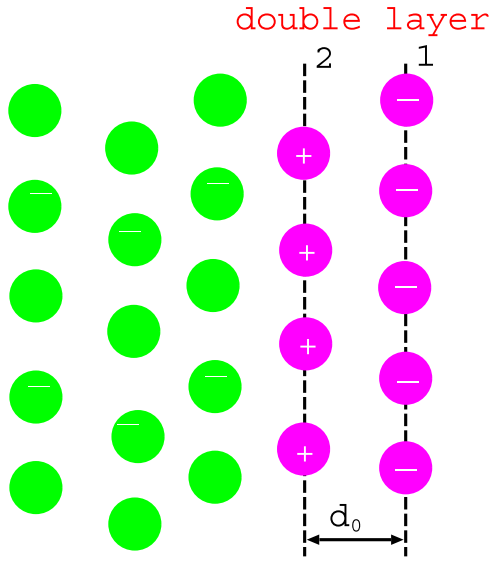


FIG. 1. Scheme of an electric double layer on a metal surface.

The potential step depends on the condition of the surface, for example, the processing method and the degree of surface contamination. If the metal is a single crystal, then the surface potential relative to its inner part depends on the distance between the lattice planes parallel to the surface. A change in the

crystallographic directions exposed on the surface of a pure polycrystalline metal leads to a change in the surface potential and the appearance of a surface charge density $\sigma(\mathbf{x})$, for which the average value $\langle \sigma(\mathbf{x}) \rangle = 0$, but $\langle \sigma^2(\mathbf{x}) \rangle \neq 0$. This is named the “patch effect,” which results in an electrostatic force of interaction between metal surfaces, which must be taken into account when measuring the Casimir force [31]. In addition to spatial fluctuations responsible for the “patch effect,” the dipole moment of the double layer will experience temporal fluctuations due to quantum and thermal fluctuations. Temporal fluctuations provide an additional contribution to the fluctuating electromagnetic field and associated fluctuation-induced electromagnetic phenomena, which include Casimir force and friction, and radiative heat transfer [7,8,32]. In addition, the interaction of the electric field with the induced dipole moment of the double layer will lead to dissipation and to additional noncontact electrostatic friction between charged surfaces. A similar mechanism (elastic response of the first few layers to an external electric field) has been discussed in the 1980s to explain the anomalous infrared absorption of metallic nanoparticles [33,34].

Consider two plates separated by a vacuum gap d . In the near-field ($d < \lambda_T = \hbar c/k_B T$) the radiative heat transfer between them is dominated by the contribution from evanescent electromagnetic waves for which the heat flux is determined by [7,8,32]

$$J^{\text{rad}} = \frac{1}{\pi^2} \int_0^\infty d\omega [\Pi_1(\omega) - \Pi_2(\omega)] \int_0^\infty k_z dk_z e^{-2k_z d} \left[\frac{\text{Im}R_{1p}(\omega, q)\text{Im}R_{2p}(\omega, q)}{|1 - e^{-2k_z d} R_{1p}(\omega, q)R_{2p}(\omega, q)|^2} + (p \rightarrow s) \right], \quad (2)$$

where

$$\Pi_i(\omega) = \frac{\hbar\omega}{e^{\hbar\omega/k_B T_i} - 1},$$

R_p and R_s are the reflection amplitudes for p and s polarized electromagnetic waves, $k_z = \sqrt{q^2 - (\omega/c)^2}$, and $q > \omega/c$ is the component of the wave vector parallel to the surface. In the presence of the electric double layer, the reflection amplitudes for the surface will not be determined by the Fresnel formulas, since the interaction of the electric field of the electromagnetic wave with the double layer will induce the surface dipole moment $p_z = \alpha_\perp E_z^+$, where α_\perp is the susceptibility of the double layer normal to the surface, and E_z^+ is the external normal component of the electric field on the surface. In the presence of the surface dipole moment the reflection amplitude for the p -polarized electromagnetic waves is determined by [15]

$$R_p = \frac{i\epsilon k_z - k'_z + 4\pi i q^2 \alpha_\perp \epsilon}{i\epsilon k_z + k'_z - 4\pi i q^2 \alpha_\perp \epsilon}. \quad (3)$$

where $k'_z = \sqrt{\epsilon(\omega/c)^2 - q^2}$, ϵ is the dielectric function of substrate. The frequency domain equation of motion of plane 1 is

$$(-\rho_d \omega^2 - i\rho_d \omega \gamma_d + K)u_1 - Ku_2 = \sigma_d E_z, \quad (4)$$

where ρ_d and γ_d are the density and damping constant for vibrations of atomic plane 1, $K = Y/d_0$ is the elastic constant for the interplanar interaction, Y is the Young modulus, d_0 is

the interplane distance, and σ_d is the surface charge density of plane 1. The displacement of surface 2 under action of stress $\sigma_2 = K(u_1 - u_2) - \sigma_d E_z$ is determined by

$$u_2 = M[K(u_1 - u_2) - \sigma_d E_z], \quad (5)$$

where M is the mechanical susceptibility that determines the surface displacement under the action of mechanical stress: $u = M\sigma^{\text{ext}}$. From Eqs. (4) and (5) we get the dipole moment induced by the electric field in the double layer

$$p_z^{\text{ind}} = \sigma_d(u_1 - u_2) = \alpha_d E_z, \quad (6)$$

where the susceptibility of the double layer is given by

$$\alpha_d = \frac{\sigma_d^2 [\omega_d^2 - (\omega^2 + i\omega\gamma_d)MK]}{K[\omega_d^2 - (\omega^2 + i\omega\gamma_d)(1 + MK)]}, \quad (7)$$

where $\omega_d = \sqrt{Y/\rho_d d_0}$. For gold $Y = 79$ GPa, the separation between (111) planes $d_0 = 2.35$ Å, the volume density $\rho = 1.9280 \times 10^4$ kgm⁻³, $\rho_d = \rho d_0 = 4.53 \times 10^{-6}$ kgm⁻², $\omega_d = 8.85 \times 10^{12}$ s⁻¹, the sound velocities for the longitudinal and transverse acoustic waves $c_l = 3240$ ms⁻¹ and $c_t = 1200$ ms⁻¹, the lattice constant $a = 4.05 \times 10^{-10}$ m, and the upper frequencies for the longitudinal and transverse acoustic waves $\omega_l^{\text{max}} = c_l \pi / a = 2.5 \times 10^{13}$ s⁻¹ and $\omega_t^{\text{max}} = c_t \pi / a = 9.3 \times 10^{12}$ s⁻¹. Because the resonant frequency ω_d lies inside the phonon band of acoustic waves, for the mechanical susceptibility it can be used for expression obtained in an elastic

continuum model [35]

$$M = \frac{i}{\rho c_t^2} \left(\frac{\omega}{c_t} \right)^2 \frac{p_t(q, \omega)}{S(q, \omega)}, \quad (8)$$

where

$$S(q, \omega) = \left[\left(\frac{\omega}{c_t} \right)^2 - 2q^2 \right]^2 + 4q^2 p_t p_l,$$

$$p_t = \left[\left(\frac{\omega}{c_t} \right)^2 - q^2 + i0 \right]^{1/2}, \quad p_l = \left[\left(\frac{\omega}{c_t} \right)^2 - q^2 + i0 \right]^{1/2}.$$

The reflection amplitude for the s -polarized electromagnetic waves is determined by

$$R_s = \frac{ik_z - k'_z}{ik_z + k'_z}. \quad (9)$$

Another heat transfer mechanism is associated with electrostatic interaction between fluctuating surface dipole moments for which the heat flux [15,16]

$$J^{ph} = \frac{1}{\pi^2} \int_0^\infty d\omega [\Pi_1(\omega) - \Pi_2(\omega)] \times \int_0^\infty dq q \frac{b^2 \text{Im}\alpha_1 \text{Im}\alpha_2}{|(1 - a\alpha_1)(1 - a\alpha_2) - b^2\alpha_1\alpha_2|^2}, \quad (10)$$

where

$$a = 4\pi q \coth qd, \quad b = \frac{4\pi q}{\sinh qd}. \quad (11)$$

For $q \gg |\varepsilon(\omega/c)|$ and $1/\varepsilon \ll 4\pi q\alpha \ll 1$ from Eq. (3) $R_p \approx 1 + 8\pi q\alpha$ and from Eqs. (2) and (10) the contribution to the heat flux from the p -polarized waves

$$J_p^{\text{rad}} \approx J^{ph} \approx J = 16 \int_0^\infty d\omega [\Pi_1(\omega) - \Pi_2(\omega)] \times \int_0^\infty dq q^3 \frac{\text{Im}\alpha_1 \text{Im}\alpha_2}{\sinh^2 qd}. \quad (12)$$

Figure 2 shows the dependence of the heat flux between two gold plates on the distance between them for different mechanisms at the temperature of one plate $T = 300$ K and the other at $T = 0$ K. For gold dielectric function [36]

$$\varepsilon = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\nu}, \quad (13)$$

where $\omega_p = 1.71 \times 10^{16} \text{ s}^{-1}$, $\nu = 4.05 \times 10^{13} \text{ s}^{-1}$. In an extreme near field according to the classical Fresnel's formulas the contribution from p -polarized waves is determined by [4]

$$S_p \approx \frac{0.2\hbar\nu^{0.5}}{cd\omega_p} \left(\frac{k_B T}{\hbar} \right)^{3.5} \quad (14)$$

and the s -wave contribution is distance independent

$$S_s \approx \frac{0.02\hbar\omega_p^2}{\mu c^2} \left(\frac{k_B T}{\hbar} \right). \quad (15)$$

Such remarkable results are consequences of singularities in the Fresnel's transmission coefficients at small q -wave vector for high conductivity materials like gold. The bulk chemical potential and work function for (111) surface of gold are

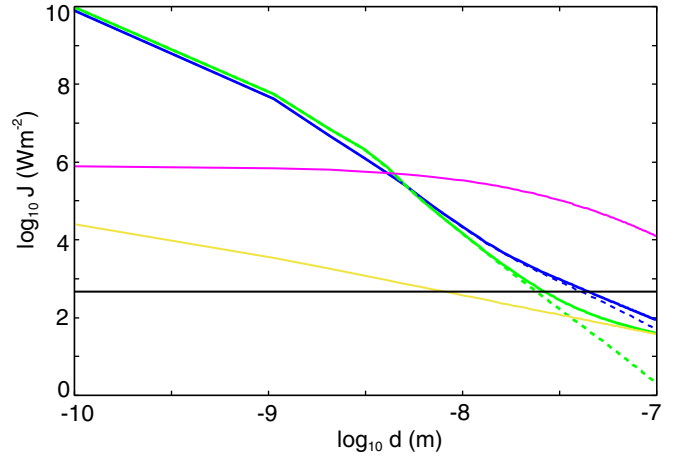


FIG. 2. The dependence of the heat flux between two plates of gold on the distance between them for different mechanisms. Temperature of one plate at $T = 300$ K, and another at $T = 0$ K. Solid and dashed blue (green) lines are for the radiative heat flux associated with p -polarized waves and electrostatic phonon heat transfer, respectively, with damping constant of the surface atomic layer vibrations $\gamma_d = 0$ ($\gamma_d = 10^{11} \text{ s}^{-1}$). Yellow line shows the radiative heat flux due to p -polarized electromagnetic waves without taking into account the double layer effect. Pink line is for the contribution from s -polarized waves. Black line for the radiative heat transfer associated with blackbody radiation.

–1 and 5.31 eV, respectively [30,37]. Thus from Eq. (1) the potential step due to the double layer $\Delta\varphi \approx 4.3$ eV. Approximating the double layer by two opposite charged planes, the plane charge density can be estimated from the relation $4\pi\sigma_d e d_0 = \Delta\varphi$ where d_0 is the separation between planes. For gold the separation between (111) planes $d_0 = 2.35 \text{ \AA}$ thus $\sigma_d \approx 0.16 \text{ Cm}^{-2}$. In contrast to the heat transfer between graphene and SiO_2 substrate which is determined by damping of flexural vibrations of graphene sheet [25], the heat transfer between gold surfaces are not sensitive to the damping constant γ_d (in Fig. 2 results for $\gamma_d = 0$ and $\gamma_d = 10^{11} \text{ s}^{-1}$ are practically identical) and is mainly determined by excitation of acoustic waves associated with coupling with double layer vibration with coupling constant M . For $d > d_c = 4.3$ nm the heat flux is dominated by practically distance independent s -wave contribution. However for $d < d_c$ the heat flux is strongly enhanced due to p -wave contribution associated with contribution from the electrical double layer, for which $S_p(d) \approx 10^{-10}/d^2$. The critical distance in heat transfer is determined by equation

$$S_p(d_c) = S_s(d_c) \quad (16)$$

from where $d_c = 4.3$ nm. The heat flux in the tip-plane configuration can be obtained from the plane-plane configuration using proximity (or Derjaguin) approximation [38]

$$P_{\text{tip}} = 2\pi \int_0^R d\rho \rho S(z(\rho)), \quad (17)$$

where R is the radius of the tip, and $S(z)$ is the heat flux between two plates separated by distance $z(\rho) = d + R - \sqrt{R^2 - \rho^2}$ denoting the tip-surface distance as a function of the distance ρ from the tip symmetry. For $R = 30$ nm, $d =$

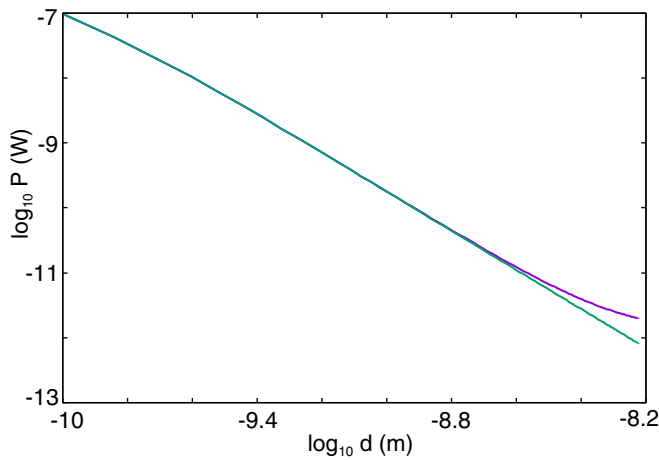


FIG. 3. The dependence of the heat flux between two gold samples on the distance between them in the tip-plate configuration. Temperatures of the tip and plate are 300 K and 0 K, respectively. Blue and green lines are for the radiative heat flux associated with p -polarized waves and electrostatic phonon heat transfer, respectively. Radius of the tip $R = 30$ nm.

3.5×10^{-10} , $n = 2$ from Eq. (17)

$$P_{\text{tip}}^{\text{max}} = 0.56\mu W \quad (18)$$

that agrees very well with the experimental data [12]. Henkel *et al.* [28] fit experimental results with equation

$$P \approx 2\pi R(d_c - d)\dot{q}. \quad (19)$$

However, in contrast to present theory, this fit does not provide microscopic explanation of physical origin of anomalously strong enhancement of heat transfer in an extreme near field.

Figure 3 shows dependence of the heat flux between gold samples in the tip-plate configuration. According to the conventional theory from Eq. (14)

$$P_p \approx \frac{0.2\pi R\hbar\nu^{0.5}}{c\omega_p} \left(\frac{k_B T}{\hbar}\right)^{3.5} \ln(R/2d). \quad (20)$$

For $R \gg d$ this contribution weakly depends on d and at $d = 1$ Å by five orders of magnitudes smaller than the contribution from electrical double layer. The maximum value $P^{\text{max}} \sim 1\mu W$ agrees with P^{max} observed in Ref. [12]. However distance dependence is stronger thus further work is needed to clarify details of this experiment. A similar situation occurs for explanation of other phenomena related to

fluctuation induced electromagnetic phenomena. Strong dependence of fluctuation induced phenomena related to gold on a structure of surface layer of gold was already noted in Ref. [39]. Experimentally it was demonstrated [40] that between charged gold tip and gold plate there is long range noncontact friction. However classical theory based on Fresnel's formulas cannot explain these results. Theory predicted friction which is by eleven orders of magnitude smaller experimental observation. In Ref. [41] a model was proposed which can explain puzzling experimental results. In this model it was proposed that surface is covered by incommensurate layer of adsorbates and within this model it was possible to explain distance dependence and magnitude for friction between gold surfaces. Thus to explain distance dependence and magnitude of heat flux in extreme near field a more sophisticated model should be developed.

The calculations of heat transfer between two plates of gold in an extreme near field were performed taking into account the electrical double layer effect. It was found that for $d < 3$ nm the double layer contribution exceeds the predictions of the conventional theory of the heat transfer by several orders of magnitudes. The presented theory predicts anomalously large nanoscale heat transfer between metals as it was observed in Refs. [12,13]. However, the origin of the anomalous heat transfer predicted by our theory and the origin of the experimental results in Refs. [12,13] can be different because experimental results were obtained in the presence of the electrostatic potential difference between the STM tip and the sample thus the potential difference can also contribute to the heat flux. However in our theory bias voltage was not assumed and predicted anomalously large heat transfer is related with the intrinsic properties of the electrical double layer on metal surface. Messina *et al.* [42] predicted that in the presence of a strong bias voltage the tunneling of electron can dominate in extreme near-field heat transfer. In contrast to these predictions, in our theory the electron tunneling was not assumed thus it proposes alternative mechanism for anomalous heat transfer in the extreme near field. It follows from the results obtained that by intentionally changing the parameters of surface dipoles with the help of engineering, one can perform the heat management at the nanoscale, which is extremely important for nanotechnology. For example, adsorbates can be used to change the surface dipole moment and enhance or attenuate heat transfer [43].

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