Electric double layer effect in an extreme near-field heat transfer between metal surfaces

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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 an electrical double layer on metal surfaces. For \( d < 3 \text{nm} \) the electrical double layer contribution exceeds the predictions of the conventional theory of the heat transfer by several orders of magnitudes. This effect is due to a coupling between the radiation electric field and the double layer dipole moment. The results obtained provide an explanation for the strong enhancement of radiative heat transfer observed in recent experiments in the extreme near field and can be used for the heat management at the nanoscale.

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All bodies are surrounded by a fluctuating electromagnetic field due to quantum and thermal fluctuations. This fluctuating electromagnetic field is responsible for the fluctuation-induced electromagnetic phenomena (FIEP) which include the Casimir forces determining the interaction between nanostructures. 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 fluctuational electrodynamics developed by Ryttov. In the framework of this theory it was theoretically predicted and experimentally confirmed, that the radiative heat flux between two bodies with different temperatures in the near field (when the distance between the bodies \( d < \lambda_T = \hbar/k_B T \) at room temperature \( \lambda_T \approx 10 \mu\text{m} \)) 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. Generally, the results of these measurements turned out to be in good agreement with the predictions based on the fluctuational electrodynamics for a wide range of materials and geometries. However, there are still significant unresolved problems in understanding heat transfer between bodies in an extreme near field (gap size \(< 10 \text{ nm} \))15,16. In Refs.15,16 the heat flux between a gold coated near-field scanning thermal microscope tip and a planar gold sample in the 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 Ryttov theory. In particular, the heat transfer observed in Ref.15 for the separations from 1 to 10 nm is orders of magnitude larger than the predictions of conventional Ryttov 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 region17,18,19–25. In Ref.18 it was shown that the heat flux in the extreme near-field can be strongly enhanced in presence of the potential difference between bodies. This enhancement is due to the electric field effect related with the fluctuating dipole moment induced on the surfaces by the potential difference. In this Communication another mechanism, related with the fluctuating dipole moment of the electrical double layer on metal surface, is considered. In contrast with the electric field effect, which is short range and operates for the separations of order \(~ 1 \text{ nm} \), the electric double layer effect is long range and operates for the separations \(< 10^2 \text{ nm} \).

The electrical 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 (Ångströms), creating a negative sheet of charges in the surface atomic plane and leaving a positive sheet of uncompensated metal ions in the sub-surface atomic planes.\(^{23,24}\) The electrical 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.\(^{20}\)

\[
A = \Delta \varphi - \bar{\mu},
\]

where \( \bar{\mu} = \mu - \bar{\varphi} \) is bulk chemical potential measured from the level of mean electrostatic potential.
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 $\sigma(x)$, for which the average value $<\sigma(x)> = 0$, but $<\sigma^2(x)> \neq 0$. 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. In addition to spatial fluctuations responsible for the “patch effect”, the dipole moment of the electrical 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 the Casimir force and friction, and radiative heat transfer.

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

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

where

$$\Pi_i(\omega) = \frac{\hbar \omega}{e^{\hbar \omega/k_B T_i} - 1}.\quad (2)$$

$R_p$ and $R_s$ are the reflection amplitudes for $p$ and $s$ polarized electromagnetic waves, $k_z = \sqrt{q^2 - (\omega/c)^2}$, $q > \omega/c$ is the component of the wave vector parallel to the surface. In the presence of the electrical 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 $\alpha \perp$ is the susceptibility of the double layer in a direction perpendicular to the surface, $E^+ + z$ 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

$$R_p = \frac{i\varepsilon k_z - k'_z + 4\pi i q^2 \alpha \perp \varepsilon}{i\varepsilon k_z + k'_z - 4\pi i q^2 \alpha \perp \varepsilon},$$

where $k'_z = \sqrt{\varepsilon(\omega/c)^2 - q^2}$. The frequency domain equation of motion of plane 1 is

$$(-\rho_d \omega^2 - i \rho_d \omega \gamma_d + K)u_1 - K u_2 = \sigma_d E^+_z,$$  

where $\rho_d$ and $\gamma_d$ 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, $\sigma_d$ is the surface...
charge density of plane 1. The interaction of the electric field with the plane 2 can be neglected due to the screening. Thus the displacement of surface 2 under action of stress $\sigma_2 = K(u_1 - u_2)$ is determined by

$$u_2 = MK(u_1 - u_2),$$

(5)

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

$$p^\text{ind}_z = \sigma_d(u_1 - u_2) = \alpha_\perp E^+_z,$$

(6)

where the susceptibility of the double layer is given by

$$\alpha_\perp = \frac{\sigma_d^2}{\rho_d[\omega_d^2 - (\omega^2 + i\omega\gamma_d)](1 + MK)},$$

(7)

where $\omega_d = \sqrt{Y/\rho_d}$.

In an elastic continuum model, $M = i\rho_c^2\pi t(\omega_c t)^2 p_l(q, \omega_c t) S(q, \omega_c t)$.

(8)

where

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

(9)

where $\rho, c_l, \text{and } c_t$ are the density of the medium, the velocity of the longitudinal and transverse acoustic waves, respectively. For $s$-polarized waves, we can neglect the interaction of an electromagnetic wave with the electrical double layer, therefore the reflection amplitude is described by the Fresnel formula

$$R_s = \frac{k_z - k_z'}{k_z + k_z'}.$$

(10)

There is also phonon heat transfer associated with electrostatic interaction between fluctuating surface dipole moments. In the electrostatic limit, the potential of the electrostatic field created in the vacuum gap between metal surfaces 1 and 2 by the surface dipole moments has the form

$$\phi(x, z) = \int \frac{d^2q}{(2\pi)^2} \left(\nu_- e^{-qz} + \nu_+ e^{qz}\right) e^{iqx}.$$

(11)

The boundary condition for the component of the electric field parallel to the surface has the form

$$E^+_q - E^-_q = iq \int_{-0}^{+0} dz E_z(z) = -iq \int_{-0}^{+0} dz \frac{dE_z}{dz} = -4\pi i q p_z.$$

(12)

Inside the metal, the strength of the electrostatic field must vanish. Whence the boundary conditions at $z = 0$

$$-4\pi p_{1z} + \nu_- + \nu_+ = 0,$$

(13)

and at $z = d$

$$4\pi p_{2z} + \nu_- e^{-qd} + \nu_+ e^{qd} = 0,$$

(14)

where $p_{iz}$ is the dipole moment for surface $i$. From (12) and (13) we get

$$\nu_+ = \frac{4\pi p_{1z} e^{-qd}}{e^{qd} - e^{-qd}} - \frac{4\pi p_{2z}}{e^{qd} - e^{-qd}}.$$
\[ \nu^- = \frac{4\pi p_{1z}e^{qd}}{e^{qd} - e^{-qd}} + \frac{4\pi p_{2z}}{e^{qd} - e^{-qd}}, \]  

(15)

\[ \phi(x, z) = \int \frac{d^2q}{(2\pi)^2} \frac{4\pi}{\sinh qd} [p_{1z}\sinh(q(d - z)) - p_{2z}\sinh qz] e^{iqz}. \]  

(16)

From [16] follows that the surface potential is related with the dipole moment per unit area: \( \varphi(x, 0) = 4\pi p_{1z}(x), \) \( \varphi(x, d) = -4\pi p_{2z}(x). \)

The electric field components for \( 0 < z < d \) have the form

\[ E_z = \frac{4\pi q}{\sinh qd} [p_{1z}\cosh(q(d - z)) + p_{2z}\cosh qz] e^{iqz}, \]  

(17)

\[ E_q = -\frac{4\pi i q}{\sinh qd} [p_{1z}\sinh(q(d - z)) - p_{2z}\sinh qz] e^{iqz}. \]  

(18)

According to the fluctuational electrodynamics[6], the surface dipole moments \( p_i = p_i^f + p_i^{ind} \) are determined by equations

\[ p_{1z} = p_{1z}^f + \alpha_1 E_1 = p_{1z}^f + \frac{4\pi q\alpha_1}{\sinh qd}(p_{1z}\cosh qd + p_{2z}), \]  

(19)

\[ p_{2z} = p_{2z}^f + \alpha_2 E_2 = p_{2z}^f + \frac{4\pi q\alpha_2}{\sinh qd}(p_{1z} + p_{2z}\cosh qd), \]  

(20)

where \( p_i^f \) is the fluctuating dipole moment due to the thermal and quantum fluctuations in solids and \( p_i^{ind} = \alpha_i E_i \) is the induced dipole moment. \( E_i = E_2(z = z_i) \) is the normal component of electric field on surface \( i = \{1, 2\} \), \( z_1 = 0 \) and \( z_2 = d \). According to the fluctuation-dissipation theorem, the spectral density of fluctuations of the surface dipole moment is determined by\[33\]

\[ \langle |p_i^f|^2 \rangle = \hbar \text{Im} \omega (\omega, q) \coth \frac{\hbar \omega}{2k_B T_i}. \]  

(21)

From (19) and (20) we get

\[ p_1 = p_1^f (1 - 4\pi q\alpha_2\cosh qd) + 4\pi q\alpha_1 p_1^f / \sinh qd \]  

\[ (1 - 4\pi q\alpha_1\cosh qd)(1 - 4\pi q\alpha_2\cosh qd) - 16\pi^2 q^2 \alpha_1 \alpha_2 / \sinh^2 qd, \]  

(22)

\[ p_2 \] is obtained from \( p_1 \) as a result of the index permutation \( (1 \leftrightarrow 2) \). The heat transfer from medium 1 to medium 2 due to interaction between fluctuating dipole moments is determined by equation\[32\]

\[ J^{ph} = 4 \int_0^\infty d\omega \int (2\pi)^2 \omega \left[ \text{Im} \omega (|E_{21}|^2) - \text{Im} \omega (|E_{12}|^2) \right] = \]  

\[ = 16 \int_0^\infty d\omega [\Pi_1(\omega) - \Pi_2(\omega)] \int_0^\infty \frac{dq^3}{\sinh^2 qd} \left[ (1 - 4\pi q\alpha_1\cosh qd)(1 - 4\pi q\alpha_2\cosh qd) - 16\pi^2 q^2 \alpha_1 \alpha_2 / \sinh^2 qd \right]^2. \]  

(23)

where \( E_{21}(E_{12}) \) is the electric field on surface 2(1) created by the fluctuating dipole moment on surface 1(2). Fig. 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 \( T = 300K \) and other at \( T = 0K \). For gold \( c_l = 3240\text{ms}^{-1}, c_t = 1200\text{ms}^{-1}, \rho = 1.9280 \times 10^4\text{kgm}^{-3}, Y = 79\text{GPa} \) and dielectric function\[34\]

\[ \varepsilon = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\nu}, \]  

(24)

where \( \omega_p = 1.71 \times 10^{16}\text{s}^{-1}, \nu = 4.05 \times 10^{13}\text{s}^{-1}, \). The bulk chemical potential and work function for (111) surface of gold are -1 and 5.31eV, respectively\[30,35\]. Thus from Eq. (1) the potential step due to the double layer \( \Delta \phi \approx 4.3\text{eV}. \)
Approximating the double layer by two opposite charged planes, the plane charge density can be estimated from the relation $4\pi \sigma_{d}d_{0} = \Delta \varphi$ where $d_{0}$ is the separation between planes. For gold the separation between (111) planes $d_{0} = 2.35\text{Å}$ thus $\sigma_{d} \approx 0.16\text{Km}^{-2}$ and the density of (111) plane $\rho_{d} = \rho d_{0} = 4.53 \times 10^{-6}\text{kgm}^{-2}$. For the damping constant for vibrations of the surface atomic plane perpendicular to the surface, an estimate was used that was obtained for the damping of the surface phonon mode (the Rayleigh waves) \[ \gamma_{d} = C \omega \] (25)

where $C = 0.17$ for gold. Eq. (25) is typical for the 2D sheet flexural vibrational damping. For example, the damping constant for the flexural phonons on graphene sheet can also be determined by Eq. (25) with $C=0.01$. On Fig. 2 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 $\gamma_{d} = 10^{11}\text{s}^{-1}$ ($\gamma_{d} = 10^{12}\text{s}^{-1}$). Red 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.

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 $T = 300 \text{K}$, and another at $T = 0 \text{K}$. 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 $\gamma_{d} = 10^{11}\text{s}^{-1}$ ($\gamma_{d} = 10^{12}\text{s}^{-1}$). Red 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.

Conclusion. The calculations of heat transfer between two plates of gold in an extreme near field were performed taking into account the double layer effect. It was found that for $d < 3\text{nm}$ the double layer contribution exceeds the predictions of the conventional theory of the heat transfer by several orders of magnitudes. Thus, the presented theory can explain anomalously large nano-scale heat transfer between metals observed in Refs. \[15,16\]. The experimental results in Refs. \[15,16\] were obtained in the presence of the electrostatic potential difference of $\sim 1\text{V}$ between the STM tip and the sample thus the electric field effect can also contribute to the heat flux. From the results presented above and in Refs. \[7,8\] follow that at the potentials difference of $\sim 1\text{V}$ the electric field effect is negligible in comparison with the double-layer effect thus the electric field effect can be excluded from explaining the experimental results in Refs. \[15,16\]. For large potential difference ($> 10\text{V}$) for subnanometer separations the contributions from the electric field effect as well as from van der Waals phonon heat transfer can be important.\[7,8\] However, these contributions are short-range in comparison with the long-range contribution from the double layer effect which dominate practically for all separations 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 produce strong enhancement of radiative heat transfer.

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