

On the analysis of thermal radiation across nanoscale gaps

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Abstract

It is shown that the analysis of radiative heat transport across a nanoscale gap must take into account the correlation between radiations from different sides of the gap. This correlation can be neglected in two cases: when the gap is considerably wider than the dominant wavelength of radiation, and when the temperatures on different sides of the gap are equal. It is further shown that the Fluctuation-Dissipation theorem used by the conventional approach to the problem does not account for the correlated components of radiations from the different sides of the gap.

Further progress in nano-technology requires efficient thermal management at the nanoscale, which is hardly possible without understanding the laws of heat transport between objects separated by a few nanometers. Since most of the heat transfer between separated objects is due to electromagnetism, the theory of radiative heat transfer [1, 2] might be expected to explain heat transfer across nanoscale gaps. The classical theory of radiative heat transport was developed in the early 1900s, with the contribution to the development of quantum mechanics as a side effect. This theory was very general in the sense that it could analyze radiative heat transport without considering specific mechanisms of thermal radiation or the contributions of specific waves. Nevertheless, the classical theory agreed well with experimental observations, and it did not need significant revisions until experiments emerged in the 1960s [3, 4] that demonstrated an unexpectedly high heat transport coefficient between bodies separated by less than a few microns. The first explanations [5] of these experiments were soon developed into the now conventional approach to radiative heat transport across micro and nanoscale vacuum gaps [6]. The predictions of this approach could be made to match the experiments from [3, 4] but, unlike the classical theory, it needed to incorporate specific assumptions about the sources of thermally excited radiation and it traced contributions of individual waves. As a result, the current conventional theory of nanoscale radiative heat transport from [6] is not as universal as the classical theory, and so it requires adjustments to particular structures. This situation suggests that this theory of nanoscale heat radiation is not completely understood, and it may have flaws that need to be identified and corrected.

To discuss the main features of radiative heat transport between bodies it suffices to consider a simple structure consisting of two half-spaces separated by a vacuum gap as was done in [6, 7] and as shown in Fig. 1.

Both the classical and the contemporary approach to radiative heat transport are based on the assumption that thermal radiations from the bodies are additive, so that the process of heat exchange is calculated as described in [6]:

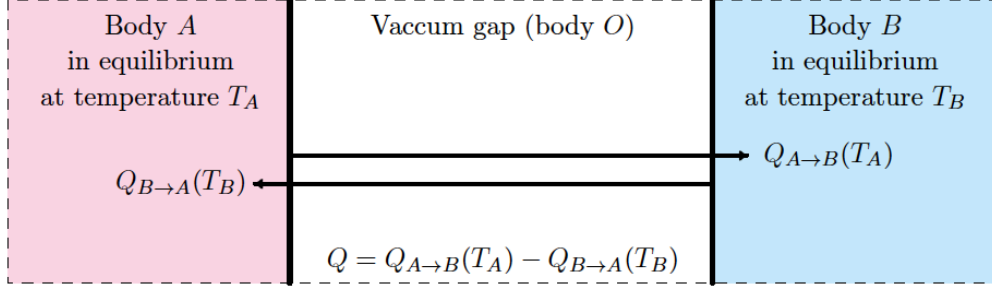


Figure 1: The conventional approach to radiative heat transport

“We first consider the fields set up by the noise sources in the medium at $z < 0$ and calculate the (averaged) Poynting vector in the z direction at the point d_+ , i.e. just inside the second medium. This is the heat transferred to the second medium due to thermal radiation from the first. In the same way we calculate the Poynting vector in the $-z$ direction at $z = 0_-$, due to sources in the second medium. The difference of the two expressions is the net energy transfer due to the temperature difference of the two bodies.” [6, Sec.V].

This implies that the net heat flux between bodies A and B from Fig. 1 can be represented as

$$Q = Q_{A \rightarrow B} - Q_{B \rightarrow A}, \quad (1)$$

where $Q_{A \rightarrow B}$ is the flux originated in A and delivered to B , while $Q_{B \rightarrow A}$ is the flux originated in B and delivered to A . The task of computing $Q_{A \rightarrow B}$ and $Q_{B \rightarrow A}$ is simplified by another two assumptions: that the radiations from different bodies are uncorrelated, as described in [7]

“Since the sources of the thermal fluctuations in media 1 and 2 are statistically independent, both fields are incoherent, and it is this which causes the additivity of fluxes ...” [7, Sec.2],

and that the bodies A and B radiate heat as if they remain in (local) equilibrium at constant temperatures T_A and T_B , respectively, [8, 9]. Then, $Q_{A \rightarrow B}$ is determined by T_A and is independent of T_B , while $Q_{B \rightarrow A}$ depends on T_B but is independent of T_A , so that

$$Q_{A \rightarrow B} \equiv Q_{A \rightarrow B}(T_A), \quad Q_{B \rightarrow A} \equiv Q_{B \rightarrow A}(T_B). \quad (2)$$

In the classical theory of heat radiation dealing with larger scales this approach leads to the Stefan-Boltzman formulas $Q_{X \rightarrow Y}(T_X) = \gamma \sigma T_X^4$, etc., resulting in the expression

$$Q \equiv Q(T_A, T_B) = \gamma \sigma (T_A^4 - T_B^4), \quad (3)$$

where σ is a universal constant and γ is a constant characterizing the absorption by the bodies. This implies that the heat transport coefficient of a gap between half-spaces has a finite value

$$\kappa(T_B) = \left. \frac{\partial Q(T_A, T_B)}{\partial T_A} \right|_{T_A=T_B} = 3\gamma \sigma T_B^3. \quad (4)$$

Since (4) does not depend on the width H of the gap this formula does not agree with the observations that the heat transport coefficient increases as H decreases below the dominant wavelength

of thermal radiation [3, 4, 10]. However, this result has been validated by so many experiments at larger scales that the assumptions behind it appear to be justified, and so its failure at nanoscale separations H is further assumed to be caused by an inaccurate evaluation of the fluxes $Q_{X \rightarrow Y}(T_X)$, which is corrected by introducing evanescent waves and tunneling effects in the contemporary theories, such as those presented in [6, 7, 11], for example.

Nevertheless, a closer look reveals that the assumptions regarding the independence and additivity of radiations from different bodies cannot be valid in cases when the distance H between the bodies approaches zero. Indeed, if the distance is smaller than the dominant wavelength of radiation, then the interaction of any two atoms on different sides of the gap with radiation can be understood by the simple explanation in the well-known undergraduate physics text [12]:

“Consider what would happen if, instead of a single atom, we had an agglomerate of atoms, say two, very close together compared with the wavelength of the light. Remember, atoms are only an angstrom or so across, while the wavelength of light is some 5000 angstroms, so when they form a clump, a few atoms together, they can be very close together compared with the wavelength of light. Then, when the electric field acts, *both of the atoms will move together*. The electric field that is scattered will then be the sum of the two electric fields in phase. . . . Our argument that the phases are independent is based on the assumption that there is a real and large difference in phase between any two atoms, which is true only if they are several wavelengths apart and randomly spaced, or moving. But if they are right next to each other, they necessarily scatter in phase, and they have a coherent interference which produces an increase in scattering.” [12, Lecture 32].

This implies that if $H < \lambda$, then the assumption about statistical independence of the radiations from different bodies is violated and, therefore, that any approach to the analysis of nanoscale radiative heat transport which uses the expression (1) cannot be correct. Moreover, since the Poynting vector is a bilinear function of the electromagnetic field, it is not additive in the sense that the energy flux carried by a superposition of two fields is not always equal to the sum of fluxes carried by individual fields. This implies, that the basic decomposition (1) may not be unconditionally valid, as suggested by the quote from [6, Sec.V].

Despite the transparency of the above reasoning there is a common misconception [6, 11, 13, 14] that the conventional approach to radiative heat transport at the nanoscale is rigorously justified by the Fluctuation-Dissipation theorem which appears as the centerpiece of the description of thermal radiation in terms of Fluctuational Electrodynamics [8, 15, 16].

Fluctuational Electrodynamics is a phenomenological theory based on an assumption that thermal radiation from a domain filled by an absorbing material is generated by stochastic extraneous currents, whose statistical distributions obey certain restrictions, which will be discussed below. These currents generate a fluctuating electromagnetic field $(\mathbf{E}(\mathbf{r}, t), \mathbf{H}(\mathbf{r}, t))$ which carries the heat flux determined by the averaged Poynting vector

$$\mathbf{S}(\mathbf{r}) = \frac{1}{2} \text{Re} \left\langle \mathbf{E}(\mathbf{r}, t) \times \overline{\mathbf{H}(\mathbf{r}, t)} \right\rangle, \quad (5)$$

where the overbar denotes the complex conjugation.

Consider the structure shown in Fig. 1, where the space V consists of two material half-spaces A and B separated by a vacuum layer O . Due to linearity, the total electromagnetic field (\mathbf{E}, \mathbf{H}) radiated by fluctuating currents in the both domains can be represented as

$$\mathbf{E} = \mathbf{E}^A + \mathbf{E}^B, \quad \mathbf{H} = \mathbf{H}^A + \mathbf{H}^B, \quad (6)$$

where $(\mathbf{E}^A, \mathbf{H}^A)$ and $(\mathbf{E}^B, \mathbf{H}^B)$ are the electromagnetic fields radiated by the domains A and B , respectively. Therefore, the total heat flux at a point \mathbf{r} in the structure is determined as the sum

$$\mathbf{S}(\mathbf{r}) = \mathbf{S}(\mathbf{r}; A, A) + \mathbf{S}(\mathbf{r}; B, B) + \mathbf{S}(\mathbf{r}; A, B) + \mathbf{S}(\mathbf{r}; B, A), \quad (7)$$

where the term

$$\mathbf{S}(\mathbf{r}, X, Y) = \frac{1}{2} \text{Re} \left\langle \mathbf{E}^X(\mathbf{r}, t) \times \overline{\mathbf{H}^Y(\mathbf{r}, t)} \right\rangle, \quad (8)$$

represents the averaged energy flux at the point \mathbf{r} caused by the combination of the electric field originating in the domain X with the magnetic field originating from the domain Y .

To make the computation of the fluxes (8) more transparent we follow [11] and limit ourselves to the case of non-magnetic media where thermal radiation is considered to be generated by an electric current with the spectrum $\mathbf{J}(\mathbf{r}, \omega)$. Then, using classical electrodymanics we find that the electric field $\mathbf{E}^X(\mathbf{r}, t)$ generated by the current in a domain X can be represented as

$$\mathbf{E}^X(\mathbf{r}, t) = \int_X d\mathbf{x} \int_0^\infty \mathbb{G}^e(\mathbf{r}, \mathbf{x}, \omega) \mathbf{J}(\mathbf{x}, \omega) e^{i\omega t} d\omega, \quad (9)$$

where $\mathbb{G}^e(\mathbf{r}, \mathbf{x}, \omega)$ is a deterministic matrix defined by the Green's functions of the considered structure and is unrelated to thermal processes. Similarly, the magnetic field $\mathbf{H}(\mathbf{r}, t)$ excited by the current in a domain Y can be represented as

$$\mathbf{H}^Y(\mathbf{r}, t) = \int_Y d\mathbf{y} \int_0^\infty \mathbb{G}^h(\mathbf{r}, \mathbf{y}, \omega) \mathbf{J}(\mathbf{y}, \omega) e^{i\omega t} d\omega, \quad (10)$$

where $\mathbb{G}^h(\mathbf{r}, \mathbf{x}, \omega)$ is another deterministic matrix.

Let E_k , H_k and S_k be Cartesian components of \mathbf{E} , \mathbf{H} and \mathbf{S} . Then

$$S_k(\mathbf{r}; X, Y) = \frac{1}{2} \text{Re} \left\langle \sum_{m,n} \epsilon_{kmn} E_m(\mathbf{r}, t; X) \overline{H_n(\mathbf{r}, t; Y)} \right\rangle, \quad (11)$$

where ϵ_{kmn} is the Levi-Civita symbol, and

$$E_m(\mathbf{r}, t) = \int_X d\mathbf{x} \int_0^\infty \sum_{p=x,y,z} G_{mp}^e(\mathbf{r}, \mathbf{x}, \omega) J_p(\mathbf{x}, \omega) e^{i\omega t} d\omega, \quad (12)$$

$$H_n(\mathbf{r}, t) = \int_Y d\mathbf{y} \int_0^\infty \sum_{q=x,y,z} G_{nq}^h(\mathbf{r}, \mathbf{y}, \omega) J_q(\mathbf{y}, \omega) e^{i\omega t} d\omega, \quad (13)$$

where J_p are Cartesian components of the vector \mathbf{J} , while G_{mp}^e and G_{nq}^h are the elements of the matrices \mathbb{G}^e and \mathbb{G}^h . These give

$$S_k(\mathbf{r}) = \text{Re} \sum_{p,q} \int_X d\mathbf{x} \int_Y d\mathbf{y} \int_0^\infty K_{kpq}(\mathbf{r}, \mathbf{x}, \mathbf{y}, \omega) \left\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \right\rangle d\omega, \quad (14)$$

where

$$K_{kppq}(\mathbf{r}, \mathbf{x}, \mathbf{y}, \omega) = \frac{1}{2} \sum_{m,n} \epsilon_{kmn} G_{mp}^e(\mathbf{r}, \mathbf{x}, \omega) \overline{G_{nq}^h(\mathbf{r}, \mathbf{y}, \omega)}, \quad (15)$$

is another deterministic tensor.

Although the spectra $J_p(\mathbf{x}, \omega)$ and $J_q(\mathbf{y}, \omega)$ are unknown, the averages $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle$, under certain conditions, can be computed by the Fluctuation-Dissipation theorem [8, 9, 15, 17], which states that **if the domain V is maintained at thermal equilibrium at uniform temperature T , then for all $\mathbf{x} \in V$, $\mathbf{y} \in V$**

$$\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle = C\omega \operatorname{Im}[\epsilon(\omega)] \Theta(\omega, T) \delta_{pq} \delta(\mathbf{x} - \mathbf{y}), \quad \mathbf{x}, \mathbf{y} \in V, \quad (16)$$

where $\epsilon(\omega)$ is the relative dielectric parameter of the medium, C is a universal real-valued constant, δ_{pq} and $\delta(\mathbf{x})$ are the standard δ -functions, and $\Theta(\omega, T)$ is the Planck's function representing the average energy of an oscillator at frequency ω in an equilibrium ensemble at temperature T .

Modern methods of electrodynamics make it possible to compute the matrix functions \mathbb{G}^e and \mathbb{G}^h of virtually any domain. This implies that if all averages $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle$ can be determined by (16), then formulas (7), (14) define the heat flux in the considered structure. However, it is shown below that despite a popular belief the described approach does not provide a way to compute the heat transport between bodies at different temperatures separated by a nanoscale vacuum gap.

Consider first the ‘‘diagonal’’ terms $\mathbf{S}(\mathbf{r}, A, A)$ and $\mathbf{S}(\mathbf{r}, B, B)$ defined by formulas (8) where the integrations by \mathbf{x} and \mathbf{y} are extended over the same domain, either A or B .

Let each of the domains A and B be in (local) equilibrium at the temperature T_A and T_B , respectively. Then, the averages $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle$ with both \mathbf{x} and \mathbf{y} in the domain X , either $X = A$ or $X = B$, can be evaluated by (16) with $T = T_X$, so that the corresponding expressions from (7) reduce to the form

$$S_k(\mathbf{r}; X, X) = C \operatorname{Re} \sum_p \int_X d\mathbf{x} \int_0^\infty K_{kpp}(\mathbf{r}, \mathbf{x}, \mathbf{x}, \omega) \operatorname{Im}[\epsilon(\omega)] \Theta(\omega, T_X) \omega d\omega, \quad (17)$$

with a completely defined right-hand side.

It is easy to see that due to symmetry of the considered structure, if the x -axis is directed orthogonal to the faces of the half-spaces, then the component $S_x(\mathbf{r}; A, A)$ from (17) can be identified with the heat flux radiated by half-space A and $S_x(\mathbf{r}, B, B)$ coincides with the part of the heat flux radiated by B . Then, introducing notation

$$Q_{A \rightarrow B}(T_A) \equiv S_x(\mathbf{r}; A, A), \quad Q_{B \rightarrow A}(T_B) \equiv -S_x(\mathbf{r}; B, B), \quad (18)$$

we re-write the x -component of (7) in the form

$$Q(\mathbf{r}) \equiv S_x(\mathbf{r}) = Q_{A \rightarrow B}(T_A) - Q_{B \rightarrow A}(T_B) + S_x(\mathbf{r}; A, B) + S_x(\mathbf{r}; B, A), \quad (19)$$

which differs from (1) by the presence of the two yet indefinite cross-terms $S_x(\mathbf{r}; X, Y)$ defined by (8) where the variables of integration \mathbf{x} and \mathbf{y} are located in not overlapping domains $X \neq Y$.

The “cross-terms” $S_x(\mathbf{r}, A, B)$ and $S_x(\mathbf{r}, B, A)$ from (19) can be easily evaluated in two special cases: when the temperatures of the domains A and B coincide, and in the classical limit when the width of the gap H considerably exceeds the dominant wavelength of thermal radiation.

Indeed, if $T_A = T_B \equiv T$, then the two domains A and B can be treated as a single domain maintained at the uniform temperature T , so that the averages $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle$ in

$$\begin{aligned} S_x(\mathbf{r}; A, B) &= \text{Re} \sum_{p,q} \int_A d\mathbf{x} \int_B d\mathbf{y} \int_0^\infty K_{xpp}(\mathbf{r}, \mathbf{x}, \mathbf{y}, \omega) \langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle d\omega, \\ S_x(\mathbf{r}; B, A) &= \text{Re} \sum_{p,q} \int_B d\mathbf{x} \int_A d\mathbf{y} \int_0^\infty K_{xpp}(\mathbf{r}, \mathbf{x}, \mathbf{y}, \omega) \langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle d\omega, \end{aligned} \quad (20)$$

can be evaluated by the formulas (16) applied to the domain $A \cup B$. However, since \mathbf{x} and \mathbf{y} in (20) are located in different domains, we see that $\delta(\mathbf{x} - \mathbf{y}) = 0$, and then (16) implies that $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle = 0$. Therefore, if $T_A = T_B$, then the cross terms (20) automatically vanish, but this information is not important because in equilibrium the net flux at any point vanishes *a priori*, so that heat transport in such cases doesn’t need to be studied.

If the width H of the vacuum layer between A and B is considerably larger than the dominant wavelength of thermal radiations, then the distance $|\mathbf{x} - \mathbf{y}|$ between points \mathbf{x} and \mathbf{y} is necessarily large because it is larger than H . It is known that the matrix elements $K_{kpq}(\mathbf{r}, \mathbf{x}, \mathbf{y}, \omega)$ include factors $\sim e^{i\gamma\omega(|\mathbf{x}-\mathbf{r}|+|\mathbf{y}-\mathbf{r}|)}$, which rapidly oscillate when the frequency ω changes. Due to these rapid oscillations the integrals in (20) tend to zeros as H increases, and when it becomes large compared to the wavelength, the integrals can be neglected, independently of the averages $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle$. So, the fast oscillations of the kernels of the integrals (20) justify the assumption of the classical and the conventional theories that bodies (separated by a distance large compared to the wavelength) radiate and scatter electromagnetic waves independently of each other.

In a more interesting case when $T_A \neq T_B$ and the width H of the gap is comparable or smaller than the dominant wavelength of thermal radiation, then the Fluctuation-Dissipation theorem cannot be applied to the joint domain $A \cup B$ because it does not have a uniform temperature. This implies that the integrals (20) cannot be evaluated and, therefore, that the expression (19) is useless because its last two terms are indefinite. Moreover, if \mathbf{x} and \mathbf{y} are located in different domains, it is not even possible to approximate the averages $\langle J_p(\mathbf{x}, \omega) \overline{J_q(\mathbf{y}, \omega)} \rangle$ by the formulas (16) with the temperature T set close to T_A and T_B , for example as $T = (T_A + T_B)/2$. Indeed, the error of such approximation will be of the order of the temperature differential $\Delta T = T_B - T_A$, which is comparable with the total heat flux Q between the domains.

If in (19) the last two terms are negligible, then (19) reduces to the representation (1) used in the classical theory of radiative heat transport, as well as in many contemporary papers, such as [6, 7, 11, 13], just to mention a few. However, while neglecting the cross-terms (20) in the classical theory is justified by the large distance between bodies, there is no justification for neglecting these terms in cases when this distance is smaller than the dominant wavelength of thermal radiation.

It must be emphasized that the inapplicability of the Fluctuation-Dissipation theorem for the analysis of the heat transport between two bodies at different temperatures is not surprising. Thus, the title “*Theory of equilibrium thermal fluctuations in electrodynamics*” of one of the most complete monographs presenting the Fluctuational Electrodynamics [17] is self-explaining. A more accessible monograph [8], which is usually referenced in the papers that apply the Fluctuation-Dissipation theorem incorrectly, explicitly states that

“... the use of equilibrium laws (including FDT) is no longer quite rigorous, but still justified if, as is often the case, the role of the transport phenomena is as yet insignificant.” [8, Page 112].

Obviously, if the transport phenomena are neglected as being insignificant then no further study is needed to conclude that the heat flux vanishes.

The above discussion implies that the conventional approach to radiative heat transport based on the Fluctuation-Dissipation theorem discards without proper justification some terms in the expression for the heat flux. However, this does not eliminate the possibility that despite the lack of a rigorous proof, the discarded terms are actually small, so that their absence only slightly affects the quantitative results and does not change the qualitative picture of radiative heat transport. To see that this is not the case, it suffices to analyze the result of [7] concerning radiative heat transport between two half-spaces filled by identical materials and separated by a vanishing vacuum gap.

Thus, Eq. 6 of [7] states that the heat flux between two half-spaces separated by a vacuum gap and maintained at temperatures T_A and T_B is represented (in our notation) by the integral

$$Q(T_A, T_B; H) = \frac{1}{\pi^2} \int_0^\infty [\Theta(\omega, T_A) - \Theta(\omega, T_B)] M(\omega; H) d\omega, \quad (21)$$

where $M(\omega; H)$ depends on the material parameters and by the width H of the gap. In particular, Eq. 14 of [7] states that for a vanishing gap between identical transparent materials

$$M(\omega, 0) = \frac{\omega^2}{4v^2}, \quad (22)$$

where v is the speed of light in the half-spaces. Then, from (21) and (22) we see that the heat transport coefficient of a fictitious zero-wide gap in a transparent medium has a finite value

$$\kappa(T_B; 0) = \frac{1}{4\pi^2 v^2} \int_0^\infty \frac{\partial \Theta(\omega, T_B)}{\partial T_B} \omega^2 d\omega, \quad (23)$$

instead of being infinite. This example shows that even if in some cases the predictions of the conventional theory of radiative heat transport may look acceptable, this theory does not eliminate even the most obvious deficiency of the classical formula (3), which is the failure to describe the heat transport across a closed and hence an imaginary vacuum gap.

It should finally be mentioned that, although the Fluctuational Electrodynamics and the Fluctuation-Dissipation theorem cannot be applied to the analysis of heat transport between closely separated bodies at different temperatures, this does not mean that they are useless. On the contrary, these theories are indispensable tools for the analysis of different phenomena related with thermal radiation in equilibrium systems, such as the Casimir forces or non contact friction between two bodies in thermal equilibrium, just to mention a few.

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