# On the applications of the Fluctuation-Dissipation theorem to nanoscale radiative heat transfer

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**Abstract**

It is shown that neither the Fluctuation-Dissipation theorem nor the conventional theory of radiative heat transport are applicable to the analysis of heat exchange between bodies separated by a nano-scale vacuum gap.

Further progress in nano-technology requires efficient thermal management at the nanoscale, which is hardly possible without understanding the laws of heat transfer between nanoscale layers and 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, but recent experiments [3] demonstrate significant deviations from the classical theory of radiative heat transfer. This suggests that this theory has flaws which show up in the nanoscale, and which need to be identified because otherwise any modification of the theory will mislead further studies in a wrong direction.

To discuss the main features of radiative heat transport in the nanoscale it suffices to consider a simple structure consisting of two homogeneous half-spaces separated by a vacuum gap, as shown in Fig. 1.

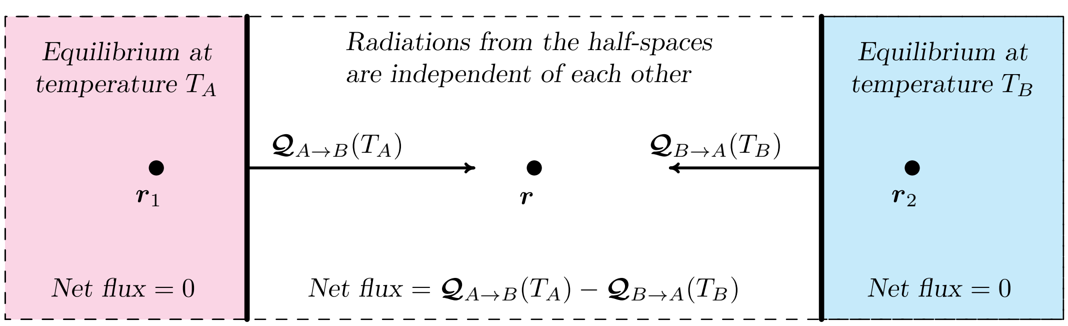


Figure 1: The conventional approach to radiative heat transport

The conventional approach to radiative heat transport is based on the assumptions that objects that emit or absorb heat may be treated as if they remain in thermal equilibrium at constant temperatures, and that the ensembles of thermally excited electromagnetic fields in heat exchanging objects are statistically uncorrelated. If these assumptions were correct then the net heat flux between bodies $A$ and $B$ could be described in the usual way as the difference

$$\boldsymbol{Q} = \boldsymbol{Q}_{A\to B}(T_A) - \boldsymbol{Q}_{B\to A}(T_B), \tag{1}$$

where the symbol $\boldsymbol{Q}_{X\to Y}(T_X)$ represents the flux, which would be radiated from the body $X$ at temperature $T_X$ to the place occupied by the body $Y$, under the assumption that $Y$ does not exist.

In larger scale systems this approach, combined with Planck's law used to determine $\boldsymbol{Q}_{A\to B}(T_A)$ and $\boldsymbol{Q}_{B\to A}(T_B)$, leads to so many well-tested results, such as the Stefan-Boltzmann law, that its assumptions appear to be well-justified by experiments. Nevertheless, a closer look suggests that these assumptions cannot be valid in cases when the distance $H$ between heat exchanging bodies is smaller than or even comparable with the dominant wavelength $\lambda$ of the thermal radiation. Indeed, the uncertainty principle applied to wave fields implies that a wave with the dominant wavelength $\lambda$ cannot be localized in a domain smaller or even comparable with $\lambda$. Therefore, if two bodies $A$ and $B$ are separated by the distance $H < \lambda$, then it is impossible to distinguish which of them radiates an electromagnetic field with the dominant wavelength $\lambda$. Correspondingly, if $H < \lambda$, then the assumption of the conventional approach regarding the statistical independence of the radiations from different bodies is violated and, therefore, this approach can not be reliably used.

Despite the transparency of the above reasoning explaining why thermal radiations from bodies separated by a nanoscale gap are correlated, there is a common misconception [4] that the statistical independence of such radiations follows from the Fluctuation-Dissipation theorem [5, 6, 7, 8], which states that under certain conditions the cross-correlation of different components of thermally excited currents in a body can be computed as

$$\langle J_l(\boldsymbol{r},\omega) J_m^*(\boldsymbol{r}_1,\omega)\rangle = \frac{1}{\pi}\epsilon_0\epsilon''(\omega)\omega\Theta(\omega,T)\delta_{lm}\delta(\boldsymbol{r}-\boldsymbol{r}_1), \tag{2}$$

where the brackets $\langle\cdot\rangle$ denote averaging, $\boldsymbol{r}$ and $\boldsymbol{r}_1$ are two points in the body, $\omega$ is the frequency of the thermal radiation, $J_l$ and $J_m$ are different components of the thermally exited electric current, $*$ represents complex conjugate, $\epsilon_0$ is the permittivity of vacuum, $\epsilon$ is the frequency-dependent relative permittivity of the matter, $\epsilon'' = \mathrm{Im}(\epsilon)$ is the imaginary part of $\epsilon$, and

$$\Theta(\omega,T) = \hbar\omega\left(\frac{1}{2} + \frac{1}{\mathrm{e}^{\hbar\omega/\kappa_B T} - 1}\right) \tag{3}$$

is the average energy of an oscillator at frequency $\omega$ in an equilibrium ensemble at temperature $T$.

It is easy to see that if (2) is valid, then the currents $J_l(\boldsymbol{r},\omega)$ and $J_m^*(\boldsymbol{r}_1,\omega)$ at the points $\boldsymbol{r}\neq\boldsymbol{r}_1$ are uncorrelated, and, correspondingly, the radiations from $\boldsymbol{r}$ and $\boldsymbol{r}_1$ are also uncorrelated. Then, assigning $\boldsymbol{r}$ and $\boldsymbol{r}_1$ to different bodies $A$ and $B$, as shown in Fig. 1, one can derive that the radiations from $A$ and $B$ are statistically independent. This suggests that the flow of energy passing from $A$ to $B$ is determined solely by the properties of the body $A$. Similarly, the flow of energy from $B$ to $A$ is determined by properties of $B$. Finally, the net flux between $A$ and $B$ can be represented as a difference of two directional fluxes as in (1).

So, while general considerations suggest that thermal radiations from slightly separated bodies may be correlated, the Fluctuation-Dissipation theorem seems to imply that they are statistically independent of each other, and that the analysis of radiative heat transfer between two bodies can be reduced to computations by the formula (1), regardless of the scale of separation. However, to be confident that the conclusion of a theorem is valid it is necessary to make sure that all of its conditions are satisfied, which, in the case considered here, is not a simple task because the Fluctuation-Dissipation theorem is rarely formulated as a mathematical statement with lists of conclusions together with conditions and assumptions. Thus, in the widely cited monographs of Rytov *et al.* [5, 6, 7, 8] some important conditions of the Fluctuation-Dissipation theorem are not explicitly included into its formulation but, instead, are discussed in physical terms and, as a result of the discussions, are included implicitly as something that may be taken for granted.

Two implicit assumptions of the Fluctuation-Dissipation theorem are obvious right from the structure of the formula (2). Indeed, since this expression involves only one temperature $T$, it is clear that the theorem is restricted to structures with a uniform temperature. Also, since (2) involves the spectrum $\Theta(\omega, T)$ of a harmonic oscillator in an equilibrium ensemble, this implies that the theorem is restricted to structures in thermal equilibrium.

When these implicit conditions are revealed, it becomes clear that the Fluctuation-Dissipation theorem is useless for the analysis of radiative heat transport between slightly separated objects. Indeed, since the entire system is supposed to be in equilibrium at temperature $T$, then, to study the structure from Fig. 1 using this theorem, it is necessary to make sure that the temperature of the half spaces are equal. But, if $T_A = T_B$, then there is nothing to study because the net flux between the half-spaces obviously vanishes.

In a more interesting case when the objects $A$ and $B$ have different temperatures $T_A \neq T_B$, the Fluctuation-Dissipation theorem can be applied to each of $A$ and $B$ separately. This means that in (2) both the points $\boldsymbol{r}$ and $\boldsymbol{r}_1$ must be located in the same domain. For example, when this theorem is applied to the domain $A$, then both $\boldsymbol{r}$ and $\boldsymbol{r}_1$ must be in $A$, and it determines the spatial correlation of thermally excited currents only inside $A$, but it gives no information about the correlation between the currents in $A$ with the currents in $B$. Similarly, if the theorem is applied to the domain $B$, then it determines the spatial correlation of the currents within $B$ but says nothing about the correlation between the currents in $A$ and $B$. As a result, the Fluctuation-Dissipation theorem, if applied correctly, does not prove the independence of thermal radiations from different domains, which is required for applications of the conventional theory of radiative heat transfer.

It must be emphasized that despite the fuzzy formulations, in [5, 6, 7, 8] the Fluctuation-Dissipation theorem is never applied in those references beyond its range of assumptions. Moreover, in [8] it is explicitly stated that this theorem is unsuitable for the analysis of radiative heat transport because it can be justified only in cases when the heat transport can be neglected:

> "*Clearly, stationary conditions are also not very difficult to achieve if the body temperature is maintained at a constant level, the energy being supplied by some external source as in the case, say, in the incandescent lamp. It is, however, clear that such a stationary case is thermodynamically not equilibrium any way, since the body energy is being converted in a one-way manner, and transport phenomena occur. (e.g. through thermal conduction). Under these conditions,* ***the use of equilibrium laws (including FDT) is no longer quite rigorous, but still justified if, as often the case, the role of the transport phenomena is as yet insignificant.***" [8, Page 112].

Obviously, if the transport phenomena is neglected as being insignificant then no further study is needed to conclude that the heat flux vanishes. Therefore, since the Fluctuation-Dissipation theorem implicitly assumes that heat transport must be neglected, it is necessary to abandon this theorem and revert instead the analysis of nanoscale radiative heat transport using the fundamental laws of thermal physics.

The above reasoning eliminates any practical need to discuss other features of the Fluctuation-Dissipation theorem prohibiting its application to the analysis of radiative heat transport in nano-scale systems. Nevertheless, it is worth mentioning that this theorem is not applicable even for the analysis of the equilibrium state of physical nano-scale systems.

Indeed, when (2) is applied to physical systems, then the $\delta$-function in this expression may not be considered in the mathematical sense, but should, instead, be considered as a function normalized as $\int \delta(\boldsymbol{r})\mathrm{d}\boldsymbol{r} = 1$ and spread over a finite domain $|\boldsymbol{r}| < \epsilon$ of a small radius $\epsilon \ll 1$, usually referred to as the correlation radius. In a real homogeneous medium, such as a large piece of a pure material, this correlation radius is of the order of intermolecular distances, which is about a few angstroms [8, 9, 3]. However, in a not perfect medium with sub-wavelength inhomogeneities:

> *"...For the sources of the fluctuational field, this gives a nonzero correlation radius which has the same order of magnitude as* ***the size of the nonlocality region in the material equations."*** *[8, Pages 122-123]*

This means that in such media the $\delta$-function in (2) should be treated as a distribution over a domain of the *"same order of magnitude as size of the nonlocality region in the material equations"*. But the system of two half-spaces from Fig. 1 has an inhomogeneity of width $H$, and this means that the carrier of the $\delta$-function in (2) is spread over the domain of the size $\sim H$ comparable with the distance between the half-spaces. Therefore, the Fluctuation-Dissipation theorem implies that the currents on different sides of the gap of the width $H$ are correlated, which violates the main assumption of the conventional theory of radiative heat transport.

It follows from the above that the net flux of thermal radiation between bodies $A$ and $B$ separated by a nanoscale gap cannot be represented by the difference (1) of uncorrelated quantities $\boldsymbol{Q}_{A\to B}(T_A)$ and $\boldsymbol{Q}_{B\to A}(T_B)$, as predicted by the conventional theory, which assumes that different bodies radiate heat independently of each other. To resolve this limitation we recently proposed a novel approach [10] where the energy flows from $A$ to $B$ and from $B$ to $A$ are represented as functions

$\boldsymbol{\mathcal{Q}}_{A\to B}(T_A, \boldsymbol{Q})$ and $\boldsymbol{\mathcal{Q}}_{B\to A}(T_B, \boldsymbol{Q})$ depending not only on the temperatures of the corresponding bodies, but also on the net flux $\boldsymbol{Q}$. Then, $\boldsymbol{Q}$ satisfies the equation

$$\boldsymbol{Q} = \boldsymbol{\mathcal{Q}}_{A\to B}(T_A, \boldsymbol{Q}) - \boldsymbol{\mathcal{Q}}_{B\to A}(T_B, \boldsymbol{Q}), \tag{4}$$

which couples the radiations from $A$ and $B$ through the unknown net flux $\boldsymbol{Q}$ and connects this flux with the temperatures $T_A$ and $T_B$. This approach is based on the extension of Planck's law of equilibrium thermal radiation to systems with a steady heat flux [11] and on the analysis of the ensembles of thermally excited electromagnetic fields in layered structures with a steady net heat flux [12]. The techniques developed in the cited papers make it possible to compute the terms in the right-hand side of (4) and, thus, to reduce the problem to the numerical analysis of equation (4). Therefore, this approach appears as a straightforward generalization of the conventional theory of radiative heat transport obtained by the elimination of the invalid assumptions about independence of thermal radiations from closely separated bodies.

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**References**

[1] M. Planck. *The theory of heat radiation.* P. Blakiston's Son & Co, 1914.

[2] R. Siegel. *Thermal Radiation Heat Transfer.* Taylor & Francis, New York, 5th edition, 2010.

[3] A. Narayanaswamy, S. Shen, L Hu, X. Chen, and G. Chen. Breakdown of the Planck blackbody radiation law at nanoscale gaps. *Applied Physics A*, 96:357–362, 2009.

[4] A. Narayanaswamy and G. Chen. Surface modes for near field thermophotovoltaics. *Applied Physics Letters*, 82(20):3544–3547, 2003.

[5] S. M. Rytov. *Theory of electric fluctuations and thermal radiation.* Academy of Science USSR, Moscow, 1953.

[6] S. M. Rytov. *Theory of electric fluctuations and thermal radiation.* Air Force Cambridge Research Center, Bedford, MA, 1959.

[7] S. M. Rytov and M. L. Levin. *Theory of equilibrium thermal fluctuations in electrodynamics.* Nauka, Moscow, 1967. (in Russian.

[8] S. M. Rytov, Yu. A. Kravtsov, and V. I. Tatatarskii. *Principles of statistical radiophysics. 3. Elements of random fields.* Springer-Verlag, Berlin-Heidelberg-New York, 1987.

[9] L. D. Landau and E. M. Lifshitz. *Electrodynamics of continuous media.* Course of Theoretical Physics, v. 8. Pergamon Press, 1960.

[10] B. V. Budaev and D. B. Bogy. Extension of Planck's law to steady heat flux across nanoscale gaps. *Applied Physics A*, 103(4):971–975, 2011. DOI 10.1007/s00339-010-6067-4.

[11] B. V. Budaev and D. B. Bogy. Extension of Planck's law of thermal radiation to systems with a steady heat flux. *Annalen der Physik*, 523(10):791–804, 2011. DOI 10.1002/andp.201100135.

[12] B. V. Budaev and D. B. Bogy. Equilibrium ensembles of electromagnetic thermal radiation in layered media. *Annalen der Physik*, 523(12):1029–1044, 2011. DOI: 10.1002/andp.201100141.