Continuous Transition of Heat Transport across a Closing Vacuum Gap from Thermal Radiation to Thermal Conduction

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Abstract— The paper discusses the common physical origin of heat conductance and heat radiation, proposes a simple model of micro and nanoscale heat transport caused by Casimir/van der Waals intermolecular forces, and addresses some difficulties arising in common applications of the conventional theory of heat radiation to micro and nano scale systems.

1. INTRODUCTION

For a long time conductance and radiation have been viewed as different mechanisms of heat transport based on different physical phenomena. Conduction dominates heat transport in solids, it is governed by the Fourier law and is described by the heat/diffusion equation. Radiation appears as the only means for heat exchange across a vacuum gap, it is governed by the laws of electromagnetism and, therefore, is described in terms of Maxwell's equations.

However, in early studies of heat transport at micro and nano scales it was noticed that the conventional theory of radiative heat transport provides calculations that differ from experimental observations. Thus, while the classical theory predicts the heat transport coefficient between two half-spaces separated by a vacuum gap should be independent of the gap's width h, it is observed [1] that this coefficient starts increasing when h decreases below a few microns, and more recent experiments [2] suggest that the increase is proportional to $1/h^2$ as the gap narrows. Although these observations contradict classical theories of radiative heat transport, they agree with common sense which suggests that as h vanishes the radiation heat transport across it should evolve to heat conductance across the resulting interface. In particular, the thermal resistance of a vacuum gap should gradually evolve to the interface thermal resistance, known as Kapitsa resistance, as the gap closes.

It is shown in [3] that when the distance h between separated bodies is comparable to the interatomic distance, then the molecules of these bodies interact via the short-range electric forces, as if they belong to a single body, and this results in heat transport by a mechanism similar to conductance. Conversely as h increases, short-range intermolecular interactions gradually diminish compared to the long-range interactions responsible for radiation, and this provides a gradual transition from heat conductance to heat radiation. It is expected that the heat conductance due to interatomic forces starts exceeding heat radiation at gaps of the order $\sim 5\,\mathrm{nm}$ and below, which suggests that this phenomenon may find application in the design of Heat Assistance Magnetic Recording (HAMR) systems where the spacing between the head and disk is less than 5 nm.

It is shown in [3] that for intermediate-range separations, between $h \sim 10\,\mathrm{nm}$ and $h \sim 3000\,\mathrm{nm}$, the heat transport coefficient follows the rate $1/h^2$ observed in [2], but not predicted by the conventional theory of thermal radiation. It is demonstrated that this failure of the conventional theory is caused by unjustified applications of some theoretical concepts, such as of the Fluctuation-Dissipation theorem, which, as stated in [4], is not applicable for the analysis of heat transport in nanoscale systems. A foundation for a self-consistent approach to radiative heat transport in nanoscale systems was proposed in [5], and a procedure for its implementation is discussed in the end of this paper.

2. A MODEL OF HEAT CONDUCTANCE DUE TO CASIMIR EFFECT

The electric field of a moving particle with the charge q is described by the expression [6]

$$\mathbf{E} = \frac{-\mathbf{q}}{\mathbf{4}\pi\epsilon}\Bigg\{\underbrace{\frac{\mathbf{e}_r}{|\mathbf{r}|^{\mathbf{2}}} + \frac{\mathbf{r}}{c}\frac{d}{dt}\left(\frac{\mathbf{e}_r}{|\mathbf{r}|^{\mathbf{2}}}\right)}_{\text{Coulomb term}} + \underbrace{\frac{1}{c^2}\frac{d^2\mathbf{e}_r}{dt^2}}_{\text{Radiation term}}\Bigg\}, \qquad \mathbf{e}_r = \frac{\mathbf{r}}{|r|}, \tag{1}$$

where c is the speed of light, and $\mathbf{r}$ is the "retarded" vector connecting the observer with the particle's position at the time $\Delta t = -|\mathbf{r}|/c$, when this field was "radiated". The first two terms in (1) describe the static Coulomb field and its correction due to the finite speed of light propagation. These terms decay proportionally to $1/|\mathbf{r}|^{\mathbf{2}}$, while the third term describing electromagnetic radiation decays proportionally to $1/|\mathbf{r}|$. Therefore, the first two terms dominate at short distances and make a major contribution to intermolecular forces which are responsible for heat conductance, and the third term dominates at long distances and is responsible for heat radiation.

Since matter consists of many particles, it may be impractical and idealistic to analyze heat transport by computing the superpositions of the fields (1) generated by all atoms. However, as in the case of conventional conductance, it is possible to derive some simple macroscopic models of heat conductance between slightly separated bodies.

It is generally accepted that heat in solid dielectrics is due to elastic waves of lattice vibrations. To model heat conductance between dielectrics separated by a vacuum gap we consider a one-dimensional chain shown in Fig. 1 where the masses $m = \rho_{\mp} a$ are located at $x_n = an < 0$ and at $x_n = h + an$, where $n \geq 0$ and $h > 0$. Assume that the springs inside the half-chains $x < 0$ and $x > h$ have the elastic moduli $\gamma_{\mp}$ and the spring connecting x_{-1} and x_0 has the modulus γ_h.

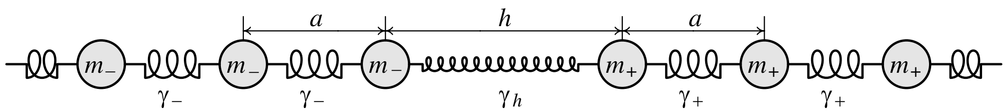


Figure 1: Two separated chains.

The motion of this chain is described by the equations

$$\rho_{\pm} a^2 \ddot{\xi}(x_n) = \gamma_{\pm} \left[\xi(x_{n+1}) + \xi(x_{n-1}) - 2\xi(x_n)\right]. \tag{2}$$

valid for all particles with $n \neq -1$, $n \neq 0$, and by two additional equations

$$\begin{aligned} \rho_- a\ddot{\xi}(0) &= \frac{\gamma_h}{h}\big[\xi(h) - \xi(0)\big] + \frac{\gamma_-}{a}\big[\xi(-a) - \xi(0)\big], \\ \rho_+ a\ddot{\xi}(h) &= \frac{\gamma_h}{h}\big[\xi(0) - \xi(h)\big] + \frac{\gamma_+}{a}\big[\xi(h+a) - \xi(h)\big], \end{aligned} \tag{3}$$

for the particles at the boundaries of the half-chains. If $a \to 0$ then (2) converges to the wave equation $\ddot{\xi}(x) = c_{\pm}^2 \nabla^2 \xi(x)$, where x is a continuous coordinate, $c_{\pm} = \sqrt{\gamma_{\pm}/\rho_{\pm}}$ are the sound speeds, and (3) reduces to the interface conditions

$$\gamma_- \xi'(0) = \gamma_+ \xi'(h) = \gamma_h \left[\xi(h) - \xi(0)\right]/h, \tag{4}$$

which compliment the wave equation describing the motions in the homogenous domains.

If the interconnection $0 < x < h$ is very strong in the sense that $\gamma_h/h \to \infty$ then (4) reduce to the condition $\xi(t,h) = \xi(t,0)$, which implies that the boundaries $x = 0$ and $x = h$ are firmly connected. In the opposite case of a very weak interconnection, (4) reduces to the Neumann conditions $\xi'(0) = \xi'(h) = 0$, which imply that the domains $x < 0$ and $x > h$ move independently of each other.

To describe a realistic three dimensional medium this model can be combined with Debye's theory, which assumes that heat is carried by acoustic waves described in terms of a pressure $p(\mathbf{r})$ related to the displacement vector field $\boldsymbol{\xi}(\mathbf{r})$ by $\rho\ddot{\boldsymbol{\xi}} = -\boldsymbol{\nabla} p$. The pressure satisfies the equations $\ddot{p} = c_{\pm}^2 \nabla^2 p$, describing the motions in the homogenous half-spaces $x > h$ and $x < 0$, and it also obeys the interface conditions

$$\gamma_- p''(0) = \gamma_+ p''(h) = \gamma_h \left[p'(h) - p'(0)\right]/h, \tag{5}$$

which generalize (4). To use these interface conditions it is necessary to know the elastic modulus γ_h of the vacuum gap of width h. If h is large compared to the intermolecular distance then γ_h can be estimated as $\gamma_h = h|F'(h)|$, where $F(h)$ is the force of interaction between the half-spaces. As shown in [7], for $h \ll \lambda_0$, where λ_0 is the dominant wavelength of electromagnetic radiation, $F(h) \approx C/h^3$. Correspondingly, the modulus γ_h has the asymptote $\gamma_h \approx \gamma_0 a^3/h^3$, where the factor $\gamma_0 a^3$ is determined by an assumption that as h reduces to the interatomic distance a, then γ_h should approach the average γ_0 of the elastic moduli of the half-spaces.

Properties of the wave motion imply that the capability of acoustic waves to carry heat between two interacting half-spaces $x < 0$ and $x > h$ is proportional to the square $|K|^2$ of the transmission coefficient K of the gap. If the width of the gap h is bigger than the the wave length Λ of thermally excited acoustic waves, which is of the order of $\sim 1\,\mathrm{nm}$ at room temperature, then

$$|K| \approx \gamma_h \Lambda/\gamma_0 h, \qquad \big(h \gg \Lambda \approx 1\,\mathrm{nm}\big), \tag{6}$$

where γ_0 is the average of the elastic moduli of the half-spaces, and γ_h is the elastic modulus of the vacuum gap. This estimate conforms with the expectations that a vanishingly narrow gap between identical media has full transmission and that a wide gap has no transmission.

The above implies that acoustic waves can penetrate the vacuum gap separating material half-spaces and that, therefore, these waves can carry heat across a vacuum gap. However, the importance of this channel of heat transfer strongly depends on the gap's width. In particular, for gaps wider than $\sim 10\,\mathrm{nm}$ electromagnetic radiation remains the sole heat carrier, but for gaps narrower than $\sim 5\,\mathrm{nm}$ acoustic waves become the dominant heat carriers.

3. RADIATIVE HEAT TRANSPORT IN NANOSCALE

Although intermolecular Casimir/van der Waals forces cause intensive heat transport across a gap narrower than $\sim 5\,\mathrm{nm}$, these forces do not provide a noticeable contribution to heat exchange across gaps wider than 30 mn, such as those experimentally studied in [2]. Correspondingly, these forces cannot explain why the heat transport coefficients of 30 nm–1000 nm gaps measured in [2] appear to be up to two orders of magnitude higher than predicted by the conventional theory.

Since experiments suggest that the theory of heat radiation has flaws which show up in the nanoscale, these flaws must be identified because otherwise any modification of the theory will mislead further studies in a wrong direction. To discuss the main flaw of this theory it suffices to consider two homogeneous half-spaces separated by a vacuum gap, as shown in Fig. 2.

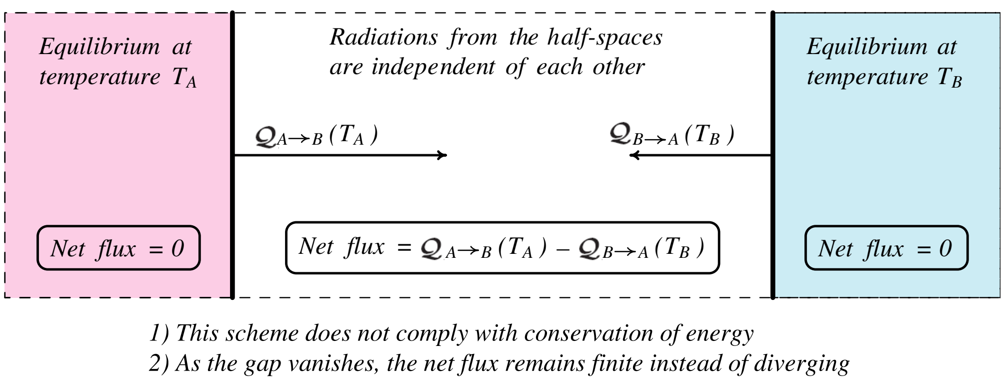


Figure 2: Inconcistency of the conventional approach to radiative heat transport.

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 represented by the difference

$$\mathbf{Q} = \mathbf{Q}_{A\to B}(T_A) - \mathbf{Q}_{B\to A}(T_B), \tag{7}$$

where the symbol $\mathbf{Q}_{X\to Y}(T_X)$ 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 $\mathbf{Q}_{X\to Y}(T_X)$, leads to many well-tested results, such as the Stefan-Boltzmann law, and hence 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 λ of the thermal radiation. Indeed, the uncertainty principle implies that a field with the dominant wavelength λ cannot be localized in a domain smaller or even comparable with λ. Therefore, if two bodies are separated by the distance $h < \lambda$, then it is impossible to distinguish which of them radiates an electromagnetic field with the dominant wavelength λ. 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 that the statistical independence of such radiations follows from the Fluctuation-Dissipation theorem [8], which states that under certain conditions the cross-correlation of different components of thermally excited currents can be computed as

$$\langle J_l(\mathbf{r},\omega)\mathbf{J}^*_{\mathbf{m}}(\mathbf{r}_1,\omega)\rangle = \frac{1}{\pi} Im(\epsilon_\omega)\omega\Theta(\omega,T)\delta_{lm}\delta(\mathbf{r}-\mathbf{r}_1), \tag{8}$$

where the brackets $\langle\cdot\rangle$ denote averaging, $\mathbf{r}$ and $\mathbf{r}_1$ are two points, ω is the frequency, J_l and J_m are the components of the thermally exited current, ϵ_ω is the permittivity, and $\Theta(\omega,T)$ is the average energy of an oscillator at frequency ω in an *equilibrium* ensemble at temperature T.

If (8) is valid, then the currents at $\mathbf{r} \neq \mathbf{r}_1$ and, correspondingly, the radiations from these points are uncorrelated. Then, assigning $\mathbf{r}$ and $\mathbf{r}_1$ to different bodies A and B, as in Fig. 2, one can show that the radiations from A and B are statistically independent. This suggests that the flow of energy from A to B is determined solely by the properties A, and the flow from B to A is determined solely by B. So, the theorem (8) seems to imply that the radiative heat transfer between two bodies is described by formula (7), regardless of the scale of separation. However, this theorem is clearly restricted to equilibrium systems where the heat flux vanishes *a priori* and, thus, does not need to be computed by any method.

To further illustrate that (8) is not applicable to the analysis of heat transport in layered nanoscale systems it suffices to notice that it involves only one temperature T, that it involves the spectrum $\Theta(\omega,T)$ of an equilibrium ensemble, and that *"... 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"*, as stated in [**?**, Page 112].

This reasoning eliminates any practical need to discuss applications of the Fluctuation-Dissipation Theorem to the analysis of radiative heat transport. Nevertheless, it is worth mentioning that this theorem is not applicable even to nanoscale systems in equilibrium.

Indeed, since (8) is applied to physical systems, then the δ-function should be considered as a distribution over a finite domain of a correlation radius $\epsilon \ll 1$, which has *"... same order of magnitude as size of the nonlocality region in the material equations"*, [**?**, Pages 122-123]. But the system of two half-spaces from Fig. 1 has an inhomogeneity of width h, and this means that the carrier of the δ-function in (8) 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 electric 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.

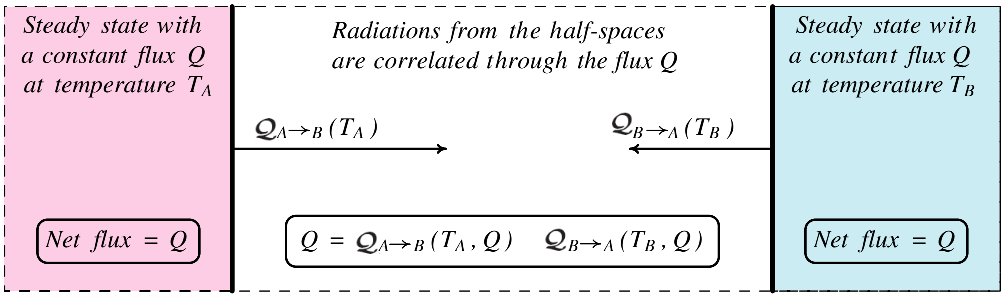
The above discussion implies that to understand heat transport across sub-micron gaps it is necessary to admit that thermal radiations from closely spaced objects are correlated and to include their correlation into the analysis, which radically distinguishes the theory of radiative heat transport in nano and low-micro scale structures from the conventional macroscopic theory.

In [4] the correlation between the radiations from closely spaced objects is taken into account by a modification of the Fluctuation-Dissipation theorem. This paper considers systems of stationary non-equilibrium, which make it possible to represent the correlations between radiations from different bodies in terms of the scattering operators associated with these bodies.

Another approach to nanoscale radiative heat transport is based on representation of the energy flows from A to B and from B to A as functions $\boldsymbol{\mathcal{Q}}_{A\to B}(T_A,\mathbf{Q})$ and $\boldsymbol{\mathcal{Q}}_{B\to A}(T_B,\mathbf{Q})$ depending not only on the temperatures of the corresponding bodies, but also on the net flux $\mathbf{Q}$. Then, $\mathbf{Q}$ satisfies the equation

$$\mathbf{Q} = \boldsymbol{\mathcal{Q}}_{A\to B}(T_A\mathbf{Q}) - \boldsymbol{\mathcal{Q}}_{B\to A}(T_B,\mathbf{Q}), \tag{9}$$

which couples the radiations from A and B through the unknown net flux $\mathbf{Q}$ and connects this flux with the temperatures T_A and T_B. This approach, illustrated in Fig. 3, is based on the extension of Planck's law of equilibrium thermal radiation to systems with a steady heat flux [5]. The technique developed on these ideas makes it possible to compute the terms in the right-hand side of (9) and, thus, to reduce the problem to the numerical analysis of that equation. Therefore, this approach appears as a straightforward generalization of the conventional theory of radiative heat transport obtained by the elimination of the invalid assumptions regarding the independence of thermal radiations from closely separated bodies.



This scheme establishes the relationship between the temperatures and the net flux

Figure 3: Self-consistency of the proposed approach to radiative heat transport.

4. CONCLUSION

The outlined method can be applied to the analysis of heat transport by any mechanism related with wave propagation, including radiative transport by electromagnetic waves and heat conduction due to acoustic waves, including that caused by Casimir/van der Waals forces. Its application to radiative transport easily explains the $\sim 1/h^2$ dependence of the heat transport coefficient of a gap of intermediate h between $\sim 10\,\text{nm}$ and $\sim 3000\,\text{nm}$. Its application to the heat conduction across the interface made it possible to get correct order-of-magnitude estimates to Kapitsa resistance, without any data fitting and with use of very crude zero-order approximations of the obtained equations. Finally, since this method describes heat radiation and heat conduction in similar terms, it provides a smooth transition between radiation and conduction across a closing vacuum gap.

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REFERENCES

1. Cravalho, E. G., C. L. Tien, and R. P. Caren, "Effect of small spacing on radiative transfer between two dielectrics," *J. Heat Transport,* Vol. 89, No. 3, 351–358, 1967.
2. Narayanaswamy, A., S. Shen, L. Hu, X. Chen, and G. Chen, "Breakdown of the Planck blackbody radiation law at nanoscale gaps," *Appl. Phys. A,* Vol. 96, 357–362, 2009.
3. Budaev, B. V. and D. B. Bogy, "On the role of acoustic waves (phonons) in equilibrium heat exchange across a vacuum gap," *APL*, Vol. 99, No. 5, 053109-1–053109-3, 2011.
4. Messina, R. and M. Antezza, "Casimir-Lifshitz force out of thermal equilibrium and heat transfer between arbitrary bodies," *EPL (Europhysics Letters)*, Vol. 95, No. 6, 61002-p1–61002-p6, 2011.
5. Budaev, B. V. and D. B. Bogy, "Extension of Planck's law of thermal radiation to systems with a steady heat flux," *Ann. der Physik*, Vol. 523, No. 10, 791–804, 2011.
6. Feynman, R. P., R. B. Leighton, and M. L. Sands, *The Feynman Lectures on Physics, V. 1,*, 3rd Edition, Pearson/Addison-Wesley, 2006.
7. Israelachvili, J., *Intermolecular and Surface Forces*, 2nd Edition, Academic Press, London, 1991.
8. Rytov, S. M., Yu. A. Kravtsov, and V. I. Tatarskii, *Principles of Statistical Radiophysics. 3. Elements of Random Fields*, Springer-Verlag, Berlin, 1987.
9. Budaev, B. V. and D. B. Bogy, "An extension of Khalatnikov's theory of Kapitsa thermal resistance," *Ann. der Physik*, Vol. 523, No. 3, 208–225, 2011.