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Classical vs generalized Kirchhoff's law in anisothermal structures

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We quantitatively analyze the thermal emission by anisothermal structures. By comparing the heat flux radiated by simple multilayered systems calculated using the classical Kirchhoff's law to the exact flux derived from fluctuational-electrodynamics theory we highlight the limits of validity of the radiometric theory to predict the thermal emission with respect to the geometric configuration, the temperature gradient and the type of materials, and highlight discrepancies as high as 50%.

Kirchhoff's law [1, 2] of thermal radiation states that a body in thermal equilibrium absorbs as much energy as it emits. Hence, by measuring the absorptivity of a body (i.e. the fraction of the energy of the incident light that is absorbed by the body) we can deduce its emissivity. This emissivity quantitatively defines the ratio between the energy emitted by this body and that of a blackbody, i.e. a perfect absorber/emitter in far-field regime, for which the emissivity is equal to 1 by definition. It is known that this law can be violated in specific situations. Hence, for instance, by breaking the optical (Lorentz) reciprocity in the system [3] it is possible to observe a deviation between the emissivity and the absorptivity. A generalization of Kirchhoff's law to systems out of thermal equilibrium was also introduced in the 50's by Rytov [4, 5] in the context of the so-called fluctuational-electrodynamics theory. This theory has been recently revisited by Greffet et al. [6] to deal with the spontaneous emission by semiconductors under optical or electrical pumping. However, despite these theoretical advances no quantitative study has been done so far to evaluate the discrepancy between the predictions made by the classical Kirchhoff's law and the rigorous calculation made in the theoretical framework of fluctuational-electrodynamics theory. Nevertheless, recently important efforts have been made to design photonic structures, including photonic crystals and metamaterials, in order to sculpt either spectrally [7–15] or directionally [16–30] their thermal emission. In these structures a notable spatial variation of temperatures can appear, making a quantitative assessment of the validity of the classical Kirchhoff's law even more relevant. In this Letter we investigate to what extent this law still remains valid in the presence of a local temperature variation. To this end we study the thermal emission of multilayer systems made with polar material or metals which are submitted to a spatial variation of temperature and we compare the predictions made from the classical Kirchhoff's law with its generalization derived from fluctuational electrodynamics.

To start let us consider the system sketched in Fig. 1 made of a slab of thickness e and of dielectric permittivity ϵ_2 placed above a semi-infinite medium of permittivity ϵ_1

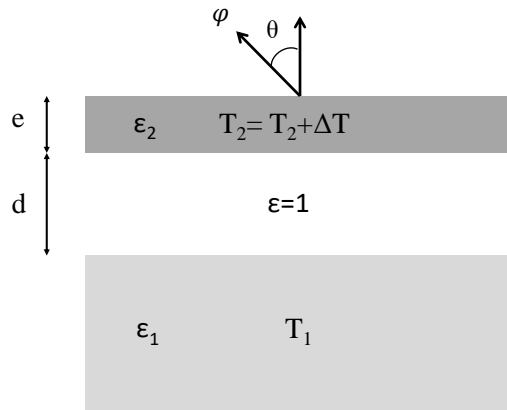


Figure 1: Schematic of a system made of a layer of thickness e and permittivity ϵ_2 above a substrate of permittivity ϵ_1 . The layer is separated from the substrate by a vacuum gap of thickness d . The substrate is held at temperature T_1 and a temperature difference ΔT is applied between the substrate and the layer. The whole system radiates a directional heat flux φ toward the surrounding environment.

and separated from it by a vacuum layer of thickness d . We now assume that the substrate is held at temperature T_1 while the slab is at temperature $T_2 = T_1 + \Delta T$. According to radiometric theory (geometrical optics) the heat flux radiated in the direction θ with respect to the normal to the surface, reads

$$\begin{aligned} \varphi_{\text{RA}}(\theta) &= 2\pi \cos \theta \sin \theta \\ &\times \frac{1}{2} \sum_{k=s,p} \int_0^\infty d\omega [\epsilon_{1,k}^{\text{dr}}(\theta, \omega) L_\omega^0(T_1) + \epsilon_{2,k}^{\text{dr}}(\theta, \omega) L_\omega^0(T_2)] \end{aligned} \quad (1)$$

where $L_\omega^0(T) = \Theta(\omega, T) \frac{\omega^2}{4\pi^2 c^2}$ is the Planck function (Θ being the mean energy of Planck oscillator) and $\epsilon_{i,k}^{\text{dr}}$ is the dressed emissivity for the polarization state $k = (s, p)$ (toward the top) of medium i in the presence of medium

$j \neq i$. These emissivities read, respectively [31],

$$\varepsilon_{1,k}^{\text{dr}}(\theta, \omega) = \frac{\varepsilon_{1,k}(\theta, \omega)T_{2,k}(\theta, \omega)}{1 - R_{1,k}(\theta, \omega)R_{2,k}(\theta, \omega)}, \quad (2)$$

and

$$\varepsilon_{2,k}^{\text{dr}}(\theta, \omega) = \varepsilon_{2,k}(\theta, \omega) + \frac{\varepsilon_{2,k}(\theta, \omega)T_{2,k}(\theta, \omega)R_{1,k}(\theta, \omega)}{1 - R_{1,k}(\theta, \omega)R_{2,k}(\theta, \omega)}, \quad (3)$$

where the presence of neighboring objects has been taken into account through Fabry-Pérot like denominator. In Eqs. (2) and (3), $R_{i,k}$ represent the intrinsic reflection coefficients of substrate ($i = 1$) and slab ($i = 2$) while $T_{2,k}$ is the intrinsic transmission coefficient of the slab in polarization k . In these expressions $\varepsilon_{i,k} = 1 - R_{i,k} - T_{i,k}$ denotes the intrinsic emissivity of medium i . Notice that the apparent emissivity used in the radiometric theory equals the average of the emissivities in the two polarizations, namely $\varepsilon_i^{\text{dr}} = (\varepsilon_{i,s}^{\text{dr}} + \varepsilon_{i,p}^{\text{dr}})/2$.

Beside this approximated result which is limited to the realm of geometrical optics a rigorous calculation of heat flux can be performed in the theoretical framework of fluctuational electrodynamics [4, 5]. In this framework the radiative heat transfer originates from the electromagnetic field produced by the random thermal motion of partial charges inside the system which behave like radiating dipoles. In this case the energy flux can be written as [32, 33]

$$\varphi_{\text{FE}}(\theta) = 2\pi \cos\theta \sin\theta \times \frac{1}{2} \sum_{k=s,p} \int_0^\infty d\omega \left[\tau_{1,k}(\theta, \omega) L_\omega^0(T_1) + [\tau_{2,k}(\theta, \omega) - \tau_{1,k}(\theta, \omega)] L_\omega^0(T_2) \right], \quad (4)$$

where $\tau_{i,k}$ are the energy transmission coefficients

$$\tau_{1,k}(\theta, \omega) = \frac{\varepsilon_{1,k}(\theta, \omega)T_{2,k}(\theta, \omega)}{|1 - \rho_{1,k}(\theta, \omega)\rho_{2,k}(\theta, \omega)e^{i2k_z d}|^2}, \quad (5)$$

$$\tau_{2,k}(\theta, \omega) = 1 - |\rho_{12,k}(\theta, \omega)|^2, \quad (6)$$

$\rho_{i,k}$ being the optical reflection coefficients of body i ($R_{i,k} = |\rho_{i,k}|^2$) which read

$$\rho_1(k, \omega, p) = r_1^p(k, \omega), \quad (7)$$

and

$$\rho_2(k, \omega, p) = \frac{r_2^p(k, \omega)(1 - e^{2ik_z d})}{1 - [r_2^p(k, \omega)]^2 e^{2ik_z d}}. \quad (8)$$

In these expressions, $r_j^{\text{TE}} = (k_z - k_{zj})/(k_z + k_{zj})$ and $r_j^{\text{TM}} = (\varepsilon_j k_z - k_{zj})/(\varepsilon_j k_z + k_{zj})$ are the Fresnel reflection coefficients of the vacuum-medium interfaces and $k_{zj} =$

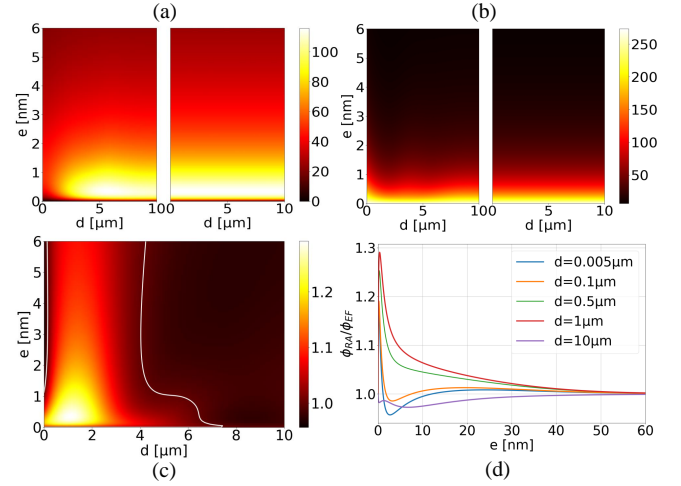


Figure 2: Thermal emission of the three-layer structure sketched in Fig. 1 and made of a gold layer on top of a SiC substrate at temperature $T_1 = 300 \text{ K}$. A temperature variation $\Delta T = 10 \text{ K}$ is applied between the substrate at the upper layer. (a) Net heat flux (in W/m^2) radiated by the layer in the presence of substrate and calculated from the radiometric (RA) and the fluctuational-electrodynamics (FE) theory. (b) Heat flux (in W/m^2) radiated by the substrate in the presence of the top layer. (c) Ratio between the net flux emitted by the whole structure using both approaches. The solid line represents the isoline $\phi_{\text{RA}}/\phi_{\text{FE}} = 1$. (d) Vertical sections of the plot shown in (c).

$\sqrt{\varepsilon_j(\omega)\omega^2/c^2 - k^2}$ is the normal component of the wave in medium j of permittivity $\varepsilon_j(\omega)$ while in the cavity it simply reads $k_z = \sqrt{\omega^2/c^2 - k^2}$, c being the speed of light. As for the reflection coefficient of block system layer/substrate it reads

$$\rho_{12,k} = \rho_{2,k} + t_{2,k}^2 \rho_{1,k} e^{2ik_z d} / (1 - \rho_{1,k} \rho_{2,k} e^{2ik_z d}), \quad (9)$$

where $t_{2,k}$ the transmission coefficient of the slab ($T_{2,k} = |t_{2,k}|^2$) and $k = \frac{\omega}{c} \sin\theta$ the parallel component of wavevector.

From Eq. (4) we can identify the apparent emissivities of substrate and slab in terms of the energy transmission coefficients as

$$\varepsilon_{1,k}^a(\theta, \omega) = \tau_{1,k}(\theta, \omega), \quad (10)$$

$$\varepsilon_{2,k}^a(\theta, \omega) = \tau_{2,k}(\theta, \omega) - \tau_{1,k}(\theta, \omega). \quad (11)$$

The comparison of Eqs. (2), (3), (10) and (11) shows the origin of differences between the radiometric approach and the electrodynamic theory. In the former, the wave nature of electromagnetic field is neglected and interference effects are not taken into account in the layer and in the vacuum cavity separating the layer from the substrate.

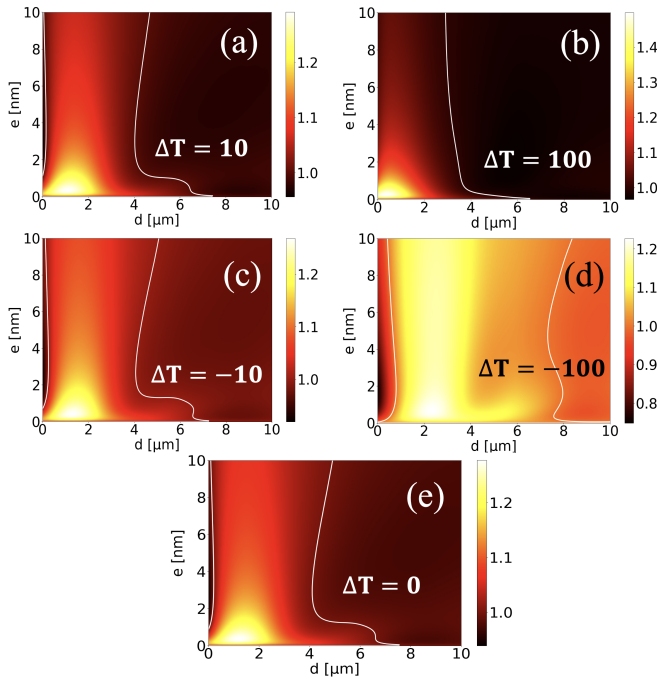


Figure 3: Ratio ϕ_{RA}/ϕ_{FE} between the predictions of the radiometry (approximated) theory and the (exact) fluctuational-electrodynamics theory for the same structure as in Fig. 2 with (a) $\Delta T = 10$ K, (b) $\Delta T = 100$ K, (c) $\Delta T = -10$ K and (d) $\Delta T = -100$ K. The solid line represents the isoline $\phi_{RA}/\phi_{FE} = 1$.

Let us now assess quantitatively, in concrete situations, the discrepancy between the thermal emission predicted by the radiometric approach and the one derived from fluctuational electrodynamics. To this end we first consider a system made of a polar substrate and a metallic film. We describe the dielectric properties of the polar material by means of a Drude-Lorentz model [34] $\epsilon(\omega) = \epsilon_\infty(\omega^2 - \omega_L^2 + i\gamma\omega)/(\omega^2 - \omega_R^2 + i\gamma\omega)$. For concrete calculations we choose $\epsilon_\infty = 6.7$, $\omega_L = 1.825 \times 10^{14}$ rad/s, $\omega_T = 1.494 \times 10^{14}$ rad/s and $\gamma = 0.9 \times 10^{12}$ rad/s, the parameters corresponding to silicon carbide (SiC). As far as the metal is concerned its dielectric permittivity is given by the Drude model $\epsilon(\omega) = 1 - \omega_p^2/\omega(\omega + i\gamma)$. For the calculations we take $\omega_p = 13.71 \times 10^{15}$ rad/s and $\gamma = 4 \times 10^{13}$ rad/s which correspond to the gold (Au) parameters [34]. We assume the substrate at temperature $T_1 = 300$ K and a temperature gradient $\Delta T = \pm 10$ K is applied between it and the layer. In Fig. 2 we plot the partial flux $\phi_{RA,FE}^{\text{slab,sub}} = \int_0^{\pi/2} d\theta \phi_{RA,FE}^{\text{slab,sub}}(\theta)$ radiated by the slab and the substrate as well as the net flux $\phi_{RA,FE} = \phi_{RA,FE}^{\text{slab}} + \phi_{RA,FE}^{\text{sub}}$ radiated by the whole structure. These fluxes are calculated by using both the radiometric approach (RA) and fluctuational-electrodynamics theory (FE).

As shown in Fig. 2(a) the radiometric approach overestimates the flux radiated by the slab at subwavelength

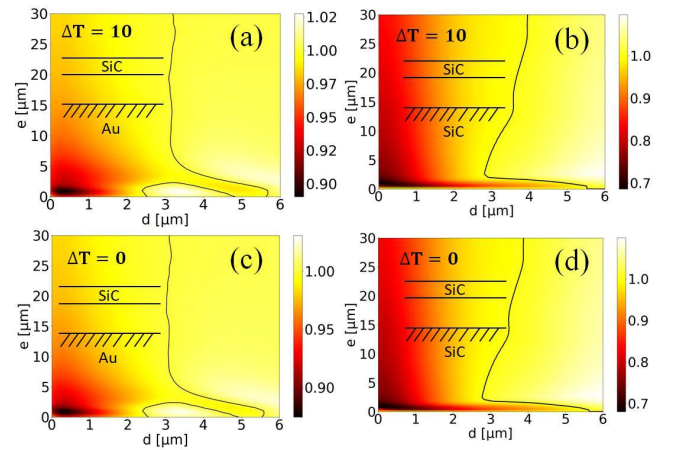


Figure 4: Ratio ϕ_{RA}/ϕ_{FE} between the predictions of the radiometry and the fluctuational-electrodynamics theory with respect to the film and cavity thicknesses for the structures shown in the insets. Same conditions as in Fig. 2. The solid line represents the isoline $\phi_{RA}/\phi_{FE} = 1$.

separation distances while the emission of substrate is approximately the same following the two approaches. In Figs. 2(c) and (d) we see that an error between 10 to 28% is observed when the thickness of the cavity is smaller than $2 \mu\text{m}$ and the film thickness is below 10 nm. Notice that these values must be taken with precaution when the film thickness is smaller than a dozen of atomic layers ($e \sim 4$ nm), the dielectric property of metal becoming size dependent below this thickness [35, 36]. Of course the observed discrepancy between the radiometric theory and the exact calculation of the thermal emission depends on the temperature difference between the substrate and the slab. The enhancement of the thermal emission as predicted by the fluctuational-electrodynamics theory (see Fig. 2) in comparison with the predictions from the radiometric theory is due to the presence of interference effects in the cavity which tend to magnify light absorption. In Fig. 3 we see that the error magnitude does not significantly depend on the sign of temperature gradient but mainly on its magnitude. Hence by comparing Figs. 3(a) and 3(c) with Figs. 3(b) and 3(d) we remark that the error is doubled when the temperature variation is increased by an order of magnitude. To highlight the purely wave effects we have added in Fig. 3 the ratio of heat flux calculations coming from the radiometric approach to the one obtained with the fluctuational-electrodynamics theory in the case where $\Delta T = 0$. The comparison of this plot with those in the presence of a temperature gradient shows that the magnitude of wave effects is weighted by the Planck functions associated to the different parts of the structure. We also plot in Fig. 3 the isoline $\phi_{RA}/\phi_{FE} = 1$ in order to identify in the space of geometric parameters where the radiometric approach fails to describe the thermal emission of system. Finally,

in Fig. 4 we show how this error is sensitive to the nature of materials. When the film is made of a polar material [Figs. 4(a) and (b)], we see that the radiometric approach, unlike the case shown in Fig. 2, underestimates the flux at subwavelength scale. This difference of behavior is due to the fact that, in this case, light is not completely screened by the film and it can penetrate inside the cavity. Then interference mechanisms can increase light absorption and therefore the overall emission of system.

In summary, in this work we have investigated to what extent the radiometric theory can be used to predict the thermal emission of anisothermal systems. Although this work was limited to simple systems it clearly demonstrates the limitations of the classical Kirchhoff's law to predict the heat flux radiated by anisothermal structures. When the temperature gradient is smaller than about ten degrees, the flux predicted by the radiometric theory is tainted by an error which is generally less than 20%. But if the temperature gradient rises to around a hundred degrees this discrepancy can become larger than 50% within simple multilayered structure. We can expect even stronger discrepancies in more complex structures where interference mechanisms can be more relevant.

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