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Size effect on the emissivity of thin films

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ABSTRACT

The size effect on the emissivity of thin films is analyzed. There are three methods for calculating film emissivity: the indirect method, the direct method based on the division of amplitude and the direct method based on Maxwell's equations combined with fluctuational electrodynamics. Traditional indirect approaches involve computation of absorption, and the emissivity is then predicted by invoking Kirchhoff's law. The direct method employed in this work, based on Maxwell's equations and fluctuational electrodynamics, does not require Kirchhoff's law. Instead, Kirchhoff's law emerges naturally from the mathematical model. A closed form expression for thickness-dependent emissivity of thin films is derived from the direct approach, and it is shown that the three existing methods lead to the same aforementioned expression. Simulation results reveal that the emissivity of metallic films increases above bulk values as the film thickness decreases. This counterintuitive behavior is due to the extraneous contributions of waves experiencing multiple reflections within the thin layer, which are usually internally absorbed for metallic bulks. Conversely, for dielectrics, the emissivity of films decreases as the film thickness decreases due to a loss of source volume. The critical thickness above which no size effect is observed for metals is about a hundred of nanometers, while it can be as large as a few centimeters for dielectrics. A simple approximate expression is finally suggested for evaluating the critical thickness; this criterion can be used as a quick reference to determine if the size effect on the emissivity of thin films should be considered.

1. Introduction

Thin films are employed in numerous engineering devices such as solar cells, optical filters and antireflection coatings [1]. In applications involving heat transfer and electromagnetic wave propagation, knowledge of the radiative properties of thin layers is crucial. Emissivity is a surface radiative property defined as the ratio of the emissive power of a surface to that of a blackbody at the same condition [2,3]. The concept of emissivity thus implies that thermal emission is a surface phenomenon. However, in reality, thermal radiation emission is a volumetric process that can often be approximated as a surface process. Waves leaving the surface of an emitting body are the result of various phenomena such as emission, absorption, transmission, and reflection. These phenomena occur throughout the entire volume of the emitter. On the other hand, only a small portion of volume below the emitting surface contributes significantly to the emitted spectrum; this small portion is defined as the critical thickness. The emissivity of a film with a thickness equal or greater than the critical thickness is referred to as the bulk emissivity. If the emitting medium is thinner than the critical thickness, the concept of emissivity, as defined in the classical theory of thermal radiation, is not valid anymore such that emissivity data reported in the literature may not be applicable to such thin layers.

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Nomenclature

c_0	speed of light in vacuum ($=2.998\times10^8$ m s^{-1})
e	electron charge ($=1.6022\times10^{-19}$ J eV^{-1})
E	wave energy ($=\hbar\omega/e$) (eV)
$\mathbf{E}$	electric field (V/m)
$\mathbf{H}$	magnetic field (A/m)
$\hbar$	reduced Planck's constant ($=1.0546\times10^{-34}$ J s)
i	complex constant ($=(-1)^{1/2}$)
$I_{b,\omega}$	spectral intensity of a blackbody (W m^{-2} sr^{-1} (rad/s)$^{-1}$)
I_ω	spectral intensity (W m^{-2} sr^{-1} (rad/s)$^{-1}$)
$\mathbf{J}^r$	stochastic current density vector (A m^{-2})
k	magnitude of wavevector ($=k'+ik''$) (rad m^{-1})
k_B	Boltzmann constant ($=1.3807\times10^{-23}$ J K^{-1})
k_ρ	parallel component of wavevector (rad m^{-1})
k_x	x-component of wavevector (rad m^{-1})
k_z	normal component of wavevector (rad m^{-1})
k_v	magnitude of wavevector in vacuum (rad m^{-1})
$\mathbf{k}$	wavevector (rad m^{-1})
q''	total, hemispherical emissive power (W m^{-2})
q''_b	total, hemispherical emissive power of a blackbody (W m^{-2})
$q''_{b,\omega}$	spectral, hemispherical emissive power of a blackbody (W m^{-2} (rad/s)$^{-1}$)
q''_ω	spectral, hemispherical emissive power (W m^{-2} (rad/s)$^{-1}$)
r_{ij}	Fresnel's reflection coefficients at interface i–j
R_j	reflection coefficient of medium j
S_z	z-component of Poynting vector (W m^{-2})
t	time (s)
t_1	film thickness (m)
t_{cr}	critical thickness (m)
t^*_{cr}	approximate critical thickness (m)
t_{ij}	Fresnel's transmission coefficients at interface i–j
T	temperature (K)
T_j	transmission coefficient of medium j

Greek symbols

α	total, hemispherical absorptivity
α'_ω	spectral, directional absorptivity
δ_λ	radiation penetration depth (m)
ε	total, hemispherical emissivity
ε_∞	high frequency dielectric constant
ε_r	dielectric function ($=\varepsilon'_r+i\varepsilon''_r$)
ε_ω	spectral, hemispherical emissivity
ε'_ω	spectral, directional emissivity
ε_0	vacuum permittivity ($=8.854\times10^{-12}$ F m^{-1})
γ	polarization state
Γ	damping factor (s^{-1})
κ	imaginary part of refractive index
λ	wavelength (m)
Θ	mean energy of a Planck oscillator (J)
ρ, θ, z	polar coordinates
σ	Stefan–Boltzmann constant ($=5.67\times10^{-8}$ W m^{-2} K^{-4})
τ'_ω	spectral, directional transmissivity
ω	angular frequency (rad s^{-1})
ω_{LO}	longitudinal optical phonon frequency (rad s^{-1})
ω_p	plasma frequency (rad s^{-1})
ω_{TO}	transverse optical phonon frequency (rad s^{-1})

In this case, a thickness-dependent film emissivity can be defined as the emissive power of the layer to that of a blackbody at the same condition.

Indirect and direct methods have been proposed in the literature for predicting the emissivity of thin films. In the indirect method, the spectral, directional absorptivity α'_ω of the layer is calculated via a conservation of energy: $\alpha'_\omega=1-\rho'_\omega-\tau'_\omega$. In this expression, ρ'_ω and τ'_ω are the spectral, directional reflectivity and spectral, directional transmissivity, respectively, calculated by considering that the layer is illuminated by an external wave with specific frequency and direction. The spectral, directional emissivity ε'_ω is then determined by invoking Kirchhoff's law, which states that the spectral, directional emissivity equals the spectral, directional absorptivity for a body in thermodynamic equilibrium [2,3]. Wang et al. [4] recently used this approach to predict the emissivity of a Fabry–Perot cavity resonator. McMahon [5] proposed a direct method where the film is sub-divided into emitting elements. Radiation emitted by each element is traced within the layer in order to determine the portion transmitted outside the film. The emissive power is then computed by integrating over the layer thickness the contribution of each element. This approach, however, treats thermal radiation as incoherent such that possible interference effects are not accounted for. Consequently, the method cannot be applied to problems where the thickness of the film is of the same order of magnitude as the radiation wavelength, since interference effects play a non-negligible role in this case. Pigeat et al. [6] generalized McMahon's approach by considering wave interferences using the division of amplitude method. The authors claim that the method is applicable for estimating the emissivity of materials with inhomogeneities of the same order of magnitude as, or greater than, the radiation wavelength. However, the method has been tested only for homogeneous media. Shih and Andrews [7] applied this method to calculate the emissivity of an oil layer on water. Wang et al. [8] employed a direct method based on Maxwell's equations and fluctuational electrodynamics to model thermal emission from a layered medium with nonuniform temperature, and they demonstrated numerically the equivalence of indirect and direct approaches. Other papers in the literature studied thermal radiation emission from complex nanostructures, such as photonic crystals [9–11].

The objective of this paper is to investigate the size effect on the emissivity of thin films using a direct approach.