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CCIT Report #820
November 2012

Electronics
Computers
Communications

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Enhanced Emission of Thermal Radiation due to Geometric Effects

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Abstract. Planck's formula for blackbody radiation was formulated subject to the assumption that the radiating body is much larger than the emitted wavelength. We demonstrate that thermal radiation exceeding Planck's law may occur in a narrow spectral range when the local radius of curvature is comparable with the wavelength of the emitted radiation. Fluctuation Dissipation Theorem needs to be employed for adequate assessment of the spectrum in this regime. Several simple examples are presented as well as experimental results demonstrating the effect.

From the early days of quantum mechanics via astrophysical measurements to today's nanostructures, blackbody radiation (BBR) played and plays a pivotal role in physics. As the emitting bodies were always much larger than the wavelength of interest, Planck's formula (PF) described adequately the general trend of the emerging radiation and any deviations were descried in terms of the so-called emissivity - which is a characteristic of the specific body or material. Conceptually, the emissivity is assumed to be always smaller than unity, explicitly assuming that PF provides the upper limit of what a body can emit. For quite some time, manufacturing techniques facilitate implementation of minute structures of a size smaller or of the same order of magnitude as the radiation wavelength, leading to a new regime of operation in which PF no longer describes adequately the BBR. Pertaining to PF as an absolute law of physics is a misconception which has been criticized even in textbooks (e.g. Ref. 1 p. 126).

Recent studies [2-5] either when optimizing thermal radiation (TR) spectrum by minimizing the reflectivity at various angles for specific wavelength ranges, or when harnessing the contribution of evanescent waves near the surface to transfer heat, push the use of PF to its limits. Moreover, when the possibility of enhanced TR spectrum in the *far-field* was claimed to be measured [6], it was ruled out as violating the second law of thermodynamics [7]. It is not our goal here to argue in favour of this particular experiment, nor of other experimental measurements of enhanced spectrum [8,9]; but rather that, in principle, this is possible due to the fact that PF is not applicable when the local radius of curvature of the body is smaller or approximately equal to the wavelength of interest.

Planck himself when determining the thermal energy density within a cavity, states that "No matter how small the frequency interval $\Delta\nu$ may be assumed to be, we can nevertheless choose l sufficiently great", where l is the cavities' dimension [Ref. 10, p.273]. Much later, Rytov [11] indicates that Planck's law is applicable only if $1 \gg \Delta\lambda/\lambda \gg (\lambda/l)^3$, "thus, the conditions for the validity of PF are first, a not too large monochromaticity of the spectral interval, and second, sufficiently large dimensions of the volume under study in comparison to λ".

Weyl [12] established a purely mathematical approach for the evaluation of high order correction terms to PF that should not be neglected for finite size cavities; others further elaborated this topic as reflected in Refs. 13-15. Baltes specifically, treated the BBR problem and summarized his work in a textbook [16]. Unlike his predecessors, Baltes calculated numerically the validity bounds of these higher-order corrections. These results have even recently been experimentally verified and reported [17].

In this Letter we aim to clarify and quantify the limits of PF. Starting from a simple rectangular closed cavity we demonstrate that in particular at small dimensions, the energy density predicted by "Planck's law" may be exceeded. We proceed to Callen's fluctuation dissipation theorem (FDT) which for the case of the impedance of a dipole emitting into free-space provides exactly PF. It is shown that if the same dipole emits within a partially open waveguide or in the vicinity of a perfectly conducting plane, the emerging energy density may exceed, in a narrow frequency range, the value predicted by PF. Subsequently, Rytov's extension of FDT is employed in order to establish the thermal radiation emitted by a body of dimensions comparable with the wavelength of interest. In this case too, values higher than those specified by PF are predicted. Finally, experimental results are presented and support our theoretical findings.

Planck's [10] original argument consists of three steps. In the *first* one he considered an ensemble of oscillators in thermal equilibrium and he established, using the classical Maxwell-Boltzmann statistics and using elemenatry quantum notions, that the energy of a system of N_{osc} oscillators at a given frequency is $\mathcal{E} = N_{\text{osc}}\Theta(T,\omega)$, wherein $\Theta(T,\omega) = \hbar\omega\left[\exp(\hbar\omega/k_B T)-1\right]^{-1}$ denotes the mean energy of a single oscillator.

The *second* step was to count the number of modes $\left(\Delta N_{\text{cavity}}\right)$ within a frequency interval –that is to say, the density of states (DoS) – in a cavity of perfectly reflecting walls of volume V_{cavity}. Subject to the tacit assumption that the wavelength is much shorter than the typical dimension of the cavity $\sqrt[3]{V_{\text{cavity}}}$, the number of modes in a range of frequencies $\Delta\omega$ starting at ω is

$$\Delta N_{\text{cavity}} = V_{\text{cavity}} \frac{\omega^2 \Delta\omega}{\pi^2 c^3}, \tag{1}$$

accounting for both possible polarizations.

His *third* step was to correlate the statistics of oscillators with the DoS in a cavity, which is a delicate matter. Essentially, there must be an equilibrium between the radiation in *vacuum* and its source in *matter*, which comes about when a wave impinging upon the walls is absorbed, causing another wave to be radiated so that the walls can be conceived as perfect reflectors; thus, the matter which is modelled by oscillators and the cavity which is modelled as a perfect resonator, must have similar oscillations. Hence , $N_{\text{osc}} = \Delta N_{\text{cavity}}$. With this assumption, the energy spectral density is

$$\frac{u}{V_{\text{cavity}}} = \frac{\omega^2}{\pi^2 c^3} \frac{\hbar\omega}{\exp\left(\hbar\omega / k_B T\right) - 1} \quad . \tag{2}$$

In the framework of this formulation, there is a distinction between the number of oscillations N_{osc} which is derived from geometrical considerations, and their mean energy Θ which is derived from statistical considerations and is therefore independent of the geometry of the problem. While Θ is correct because there is a large number of possible energy states in an harmonic oscillator, and $\mathcal{E} = N_{\text{osc}}\Theta$ is practically always correct since the number of atoms (microscopic emitters) is very large, one can question the validity of the calculation of the DoS which is only a good approximation for a cavity of "infinite" volume in respect to the wave-lengths of interest. A formal mathematical proof for the validity of (1) given this assumption, is found in Courant [18]. As already indicated, the DoS was thoroughly studied by Baltes and others [12-16] and the correction to (1) due to the finite size cavity of perfectly reflecting walls is given by [16]: $\Delta N_{\text{cavity}} / \Delta\omega = V_{\text{cavity}}\omega^2 / \pi^2 c^3 - \Lambda / 2\pi c + \ldots$, where $\Lambda_{\text{cuboid}} = a_x + a_y + a_z$, $\Lambda_{\text{sphere}} = 4R/3$ or $\Lambda_{\text{cylinder}} = 4H/3 + \pi R$.

It is evident from Eq.(2) that Planck's BBR spectrum is linearly dependent on the volume of the cavity. Essentially we demonstrate that when the typical dimensions of the emitter are smaller or comparable to the wavelength of interest, a geometric form-factor needs to be included in PF. Our starting point is to consider various cavities all of the *same volume*, but of different shapes. This is accomplished by taking a *cube* and flattening it out into a *thin film* on the one hand, or thinning it into a *rod* on the other hand. In both cases we keep the base to be a square $a_x = a_y$, as illustrated in the inset of figure 1. Our goal at this point is to establish the spectral density with special emphasis on the geometry of the cavities characterized by the parameter $a_z^2 / a_x a_y$ which is unity for a cube – corresponding to the minimum of the surface area. In the examples that follow, we push this dimensionless parameter to extremes (rod $a_z^2 / a_x a_y \gg 1$ or thin film $a_z^2 / a_x a_y \ll 1$).

In figure 1 we compare the energy density for a film, rod and cube. The various cavities store zero energy below the first mode of oscillation, and a certain amount of radiation at frequencies above that, when ignoring the first few modes for which one cannot ascribe a DoS (see top-right inset of figure 1). As these are low frequencies, there is a great deviation from Planck's law which conceptually is not valid in this range. More interesting is the deviation at higher frequencies, in which Planck's law is supposedly exact. While the cube (which has relatively large dimensions) is very close to Planck's law, the rod and film are not. The smaller the base of the rod or the height of the film, the more extreme are the deviations from Planck's law, and local enhancement of the energy spectrum may be greater than two orders of magnitude. This is one of the important results of the present study. It is interesting to compare the results of the film to the results of the discussion on heat transfer between two infinite planes which are closely spaced [19]. The latter is a 2D version of the former and thus the similarity.

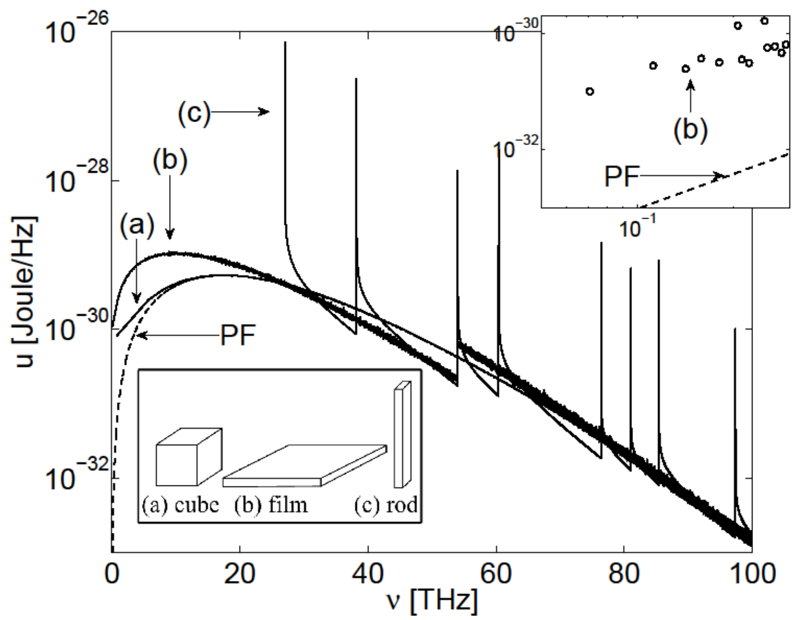

Figure 1: Comparison of enegy density within various cavities**.** Rectangular cavities of equal volume ($0.025[mm^3]$) are employed for assessing the energy density spectrum at extreme geometries: the rod (c) and film (b) have a similar surface area of $18[mm^2]$ (left-bottom inset); $T = 300[^\circ K]$. Planck's curve is plotted for comparison (dotted). For a thin rod, near cut-offs, the spectrum in a closed cavity may be orders of magnitude greater than in the case of a same volume cube (a). (top-right inset) Energy levels of the film at low frequencies, depicting the first modes of oscillation which are distinct and do not form a continuum, thus the principle of DoS is not well defined.

Another approach for the assessment of thermal radiation requires employing the so-called Fluctuation-Dissipation Theorem (FDT) developed by Callen and Welton [20].

Contrary to Planck's approach where we considered the blackbody radiation spectrum from the perspective of the *electromagnetic field*, now we investigate the spectrum from the perspective of the *oscillating electron* in the matter surrounding the cavity. The latter is assumed to be in thermodynamic equilibrium with the radiation. Denoting by $V(t)$ the effective voltage fluctuation experienced by the electron, its average is assumed to be zero $\left(\langle V\rangle = 0\right)$ and the second moment was found by Callen to be given by $\left\langle V^2\right\rangle = \dfrac{2}{\pi}\int_0^\infty d\omega R(\omega)\mathcal{E}(\omega,T)$, where $\mathcal{E} = \Theta + \hbar\omega/2$ and $R(\omega) = \mathrm{Re}\left[Z(\omega)\right]$ with $Z(\omega)$ denoting the ratio between this voltage and the reaction of the electrons (charge Q), $Z = V/\dot{Q}$; the term $\hbar\omega/2$ in $\mathcal{E}$ corresponds to vacuum fluctuations and will be ignored in what follows.

As a trivial example, Callen and Welton considered an oscillating dipole $(p = ed)$ which radiates into free-space (FS) a total average power $P_{FS} = \left(\eta_0/12\pi\right)\left(\omega^2 p/c\right)^2$. To this harmonic oscillator one may attribute a current amplitude $I = \omega p/d$ therefore, the *radiation resistance* is $R_{\mathrm{FS}}(\omega) = 2P_{FS}/|I|^2 \propto \omega^2$. Subject to these observations, the mean square of the fluctuating electric field in the vicinity of the dipole is $\left\langle E_z^2\right\rangle = \eta_0 \dfrac{1}{3\pi^2 c^2}\int_0^\infty d\omega\omega^2\mathcal{E}(\omega,T)$. Now, the relevance of FDT to BBR is evident, as the energy density experienced by the three dipoles $\left(p_x, p_y, p_z\right)$ located at a given point in free-space is $\varepsilon_0\left\langle E^2\right\rangle = \dfrac{1}{\pi^2 c^3}\int_0^\infty d\omega\omega^2 E(\omega,T)$ – which is identical to Planck's BBR formula.

This example provides us with a clear hint as of what approach should be adopted in order to exceed the value predicted by Planck's formula: we need to determine a configuration wherein the impedance experienced by the dipole is larger than that when the latter emits into free-space. In fact, the power emitted by the same dipole located at the centre ($r = 0$) of a dielectric layer medium $\left(R_{int} \le r \le R_{\text{ext}}\right)$ may exceed, near resonance, the free-space value by almost one order of magnitude – see figure 2 which shows the emitted TR spectral density for a SiC layer; the dielectric coefficient is illustrated in the inset.

FDT enables a relatively simple estimate of the geometric effect in case the latter is of the order of the wavelength. Two additional examples warrant consideration: a dipole located at a height h from an ideally conducting plane and a dipole oscillating in a partially open structure such as a half-infinite waveguide of rectangular cross-section $\left(a_x \times a_y\right)$. Based on a simple image-charge argument, one should not expect, in the first example, an enhancement of more than a factor of 2 in the emitted power. If a larger enhancement is required, it would be necessary to employ an infinite series of image-charges as is the case in the second example.

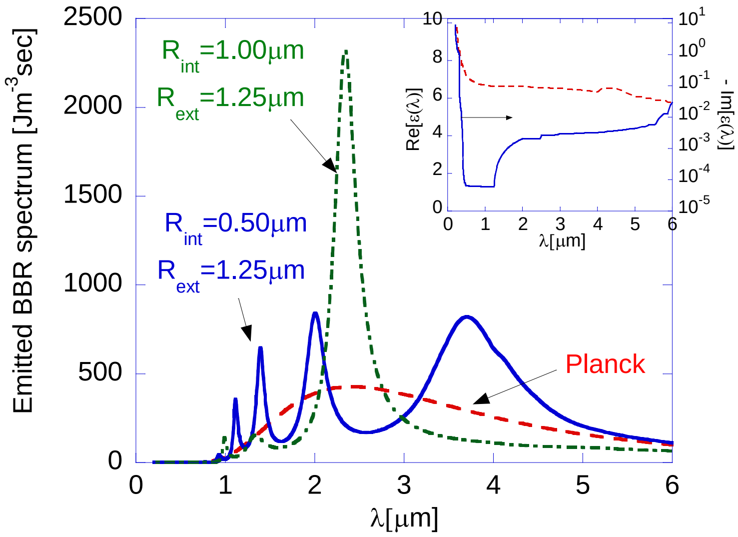

Figure 2: The emitted radiation by dipoles located in the center of a SiC dielectric layer for $R_{\text{int}} = 0.5\mu\text{m}$, $R_{\text{ext}} = 1.25\mu\text{m}$ (solid) and for $R_{\text{int}} = 1.0\mu\text{m}$, $R_{\text{ext}} = 1.25\mu\text{m}$ (dot-dash). At resonance the emitted energy spectrum may exceed the Planck's prediction for a body much larger than the wavelength of interest by almost one order of magnitude; the dashed line illustrates Planck's formula ($T = 1200[^{\circ}K]$). In the inset we specify the dielectric coefficient of SiC used in the simulation.

It is convenient to represent the emitted power in terms of the free-space value and a *form factor*, $P^{(x,y,z)} = P_{FS} F^{(x,y,z)}(\omega)$, that depends on the dipole's alignment relative to the geometry. For the first case, there is equal probability for the dipole to be alligned in a specific direction, the total form factor is $F = \left(F^{(x)} + F^{(y)} + F^{(z)}\right)/3$. All four quantities are plotted in the top frame of figure 3 and two conclusions are evident: first, for large values of $\omega h / c$ the total form-factor approaches unity; near the surface, only the perpendicular dipole contributes. Second and most important in the context of this study, the form factor may exceed unity, implying that the emitted TR spectrum may exceed the classical PF.

For the second configuration, a uniform distribution of dipoles filling up the volume $\left(a_x \times a_y \times a_z\right)$ in a semi-infinite rectangular waveguide, the average of the form factors is illustrated in the central frame of figure 3. As anticipated, for short wavelengths the form-factor always converges to unity; while for wavelengths of the same order as the waveguide's cross section, there is an enhancement of the radiation emitted, as compared to a dipole in free space. Clearly, the TR spectrum may exceed the free-space value by more than one order of magnitude.

Rytov further developed Callen's FDT [21] extending it from discrete to distributed elements by employing Lorentz reciprocity theorem. As a straight forward example we consider a sphere of radius a made of Tungsten [22]. To illustrate the form factor, the radiation intensity in the case of a finite radius is divided by that of the infinite case $(a \to \infty)$. This ratio reduces to the normalized absorption cross section $\bar{\sigma}_{\rm abs} = \sigma_{abs} / \pi a^2$ of Mie scattering [1]. It is well known that $\bar{\sigma}_{\rm abs}$ can be greater than unity, therefore it is straightforward that the thermal radiation should be greater than the classical derivation which accounts only for the geometrical area.

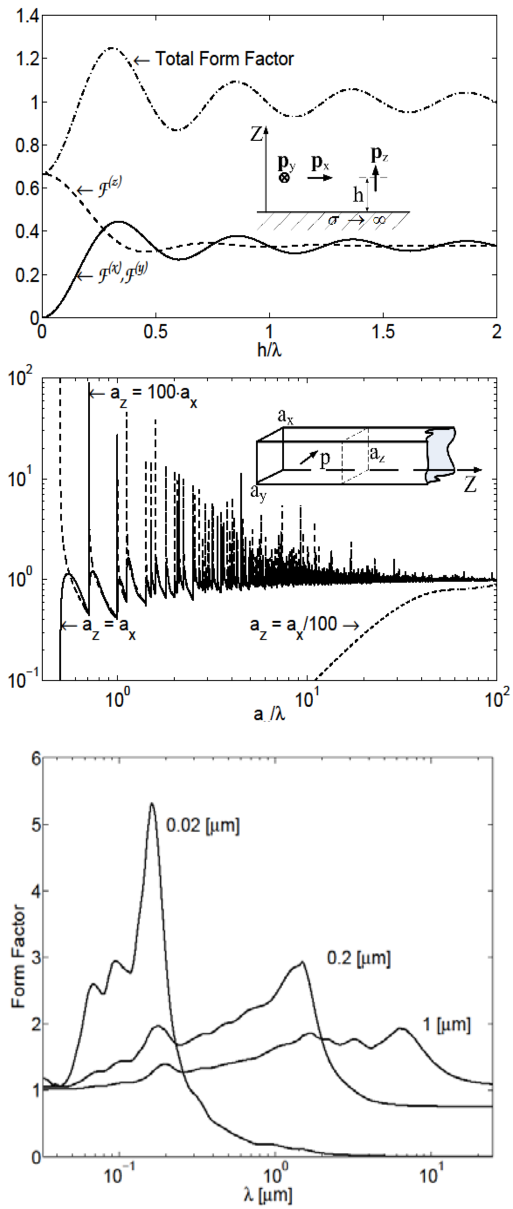

Figure 3: (top) Form factor of a dipole above an ideal plane as a function of its height normalized to the wavelength. A 20% enhancement is observed when $h/\lambda \sim 0.3$. (center) The radiation form factor for a uniform distribution of dipoles confined to the volume $a_x \times a_y \times a_z$ within a half infinite rectangular waveguide, as a function of the waveguide base normalized to the wavelength. For large a_z (dashed) the graph exhibits pronounced amplification of the thermal energy at $\lambda \sim a_x$. When $a_z = a_x$ (solid), there is a suppression of the first mode at $a_x = \lambda/2$. (bottom) Form factor of a Tungsten sphere, for various radii: 0.02, 0.2, and 1 [µm]. For wavelengths shorter than the radius the form-factor converges to unity. For wavelengths comparable with the radius the resonant character of the form factor is clearly revealed.

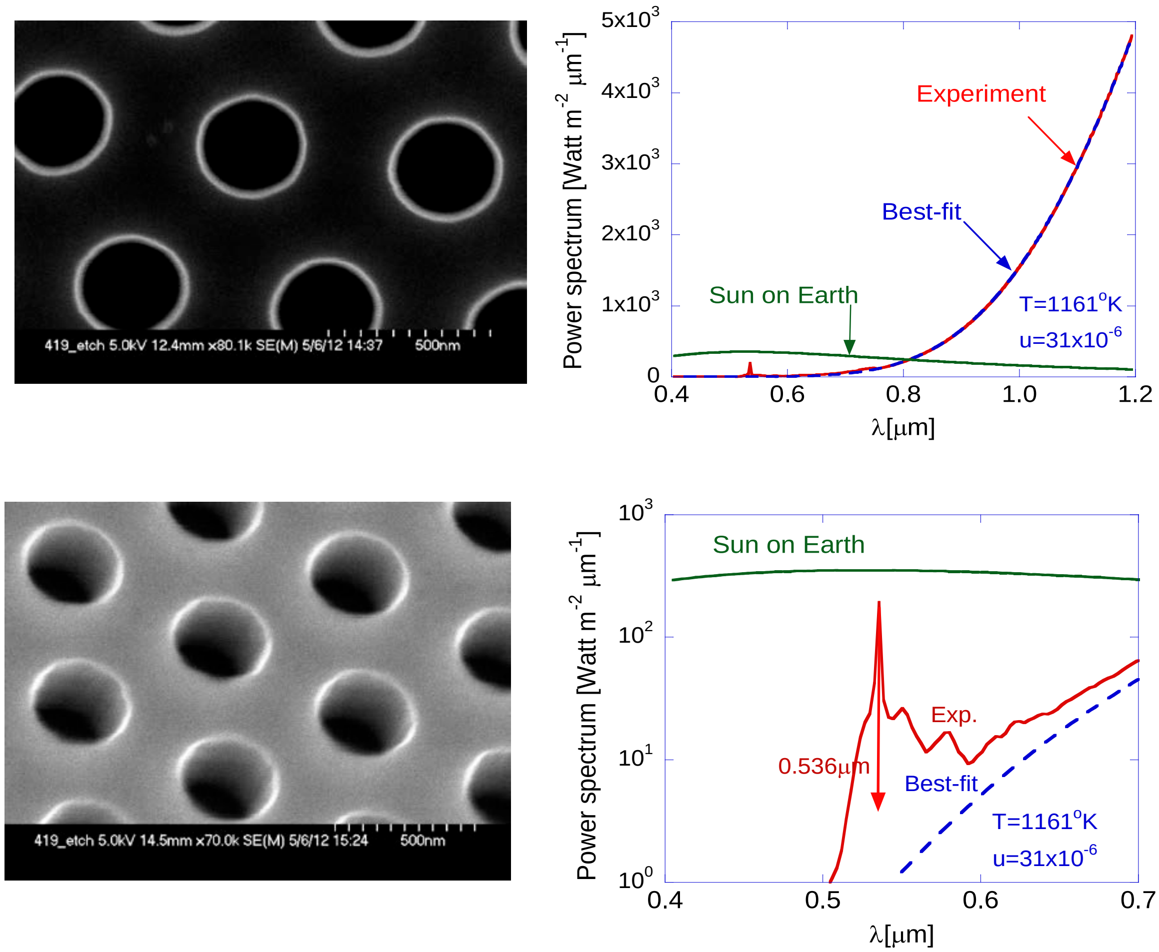

Figure 4: (left column) SEM picture of the emitting surface. Cylindrical voids (cavities) of 300[nm] diameter and a similar height, and 600[nm] pitch. (right column) Energy flux spectrum (top) as extrapolated to the surface of the blackbody (solid) and a best fit to Planck's blackbody formula (dashed). A zoom-in of the range between 0.4 – 0.7[μm] (bottom). It clearly shows that there is a significant emission enhancement. As a reference, we plot the Sun's energy-flux spectrum as measured on Earth.

Bottom frame of Figure 3 illustrates the form-factor of the sphere and it clearly reveals that its value exceeds unity for wavelengths of the the same order of magnitude as the radius (e.g. see maxima of ~ 5.5), it converges to unity at wavelengths significantly shorter than the radius.

We conducted an experiment whose goal was to demonstrate that in a narrow frequency range, thermal radiation may exceed the value predicted by PF. A set of perforated Si wafers polished with an accuracy of 1[nm] has been examined. The geometry is revealed by two SEM pictures in the left frames of figure 4. It consists of 300[nm] diameter voids with a similar height and a pitch of 600[nm]. Due to the relatively high loss, no electromagnetic coupling between the voids is expected thus no collective effect are anticipated, in other words, each void acts as a separate resonator.

After being inserted in a furnace, the wafer was gradually warmed up to about 900[^{o}C] and the thermal radiation emitted was measured by a spectrometer (CI Systems SR-5000) located about 3 meters away. Prior to measurements the spectrometer was calibrated with a standard blackbody. In the top-right frame of figure 4, the solid curve illustrates the experimental data of the energy flux spectrum as extrapolated to the surface of the blackbody. The dashed curve is a best fit to Planck's formula for the energy flux, $S\left(\lambda,T\right)d\lambda = cu\left(\omega,T\right)d\omega$. For the range between 0.4 – 1.2[μm] we found that the effective temperature is $T_{\exp} = 1161^o K$ and $\nu_{\exp} = 31.5\times10^{-6}$; these two parameters minimize the functional $\sum_i \left[S_i - \nu S\left(\lambda_i,T\right)\right]^2$ or explicitly $\nu_{\exp} = \left\langle S\left(\lambda_j,T\right)S_j\right\rangle_j / \left\langle S\left(\lambda_j,T\right)^2\right\rangle_j$ whereas

$$T_{\exp} = \min\left\{\sum_i \left[S_i - \left\langle S\left(\lambda_j,T\right)S_j\right\rangle_j \left\langle S\left(\lambda_j,T\right)^2\right\rangle_j^{-1} S\left(\lambda_i,T\right)\right]^2\right\}.$$

Each data point $\left(S_i\right)$ corresponds to the maximum value from a sample of 120 measurements at each wavelength; the wavelength resolution is 3[nm] . Except at short wavelengths, the two curves are essentially indistinguishable.

Bottom-right frame of figure 4 is a zoom-in of the range between 0.4 – 0.7[μm]. It clearly shows that there is a significant emission enhancement in particular in the range where the radiation overlaps geometric resonances. The peak occurs at 0.536[μm] and it is more than 200 times larger than the value predicted by Planck's formula at this wavelength and temperature. As a reference, we plot the Sun's energy-flux spectrum as measured on Earth.

In conclusion, the validity of Planck's black body formula is limited to geometries where the local radius of curvature of the emitting surface is larger than the radiation's wavelength. When this is not the case, Fluctuation Dissipation Theorem must be adopted for adequate assessment of the emitted radiation energy and a form-factor must be incorporated in order to properly describe the emission. In the framework of the quasi-analytic examples presented in this Letter we showed that in a narrow range of wavelengths, the emitted spectrum may exceed Planck's value by orders of magnitude in closed structures or by almost two orders of magnitude in open ones. Experimental results support this conclusion.

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Acknowledgement

LS acknowledges the help of O. Nadir and U. Avigad from SolariPhy Ltd. as well as that of A. Gil and I. Shapiro of CI Systems with the experimental set-up and measurements.