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Revisiting thermal radiation in the near field

*Le rayonnement thermique revisité en champ proche*

Jean-Jacques Greffet

Laboratoire Charles-Fabry, Institut d'Optique Graduate School, CNRS, Université Paris-Saclay, 2, avenue Fresnel, 91127 Palaiseau, France

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ABSTRACT

Thermal radiation is generally assumed to be both spatially and temporally incoherent. In this paper, we challenge this idea. It is possible to design incandescent sources that are directional and spectrally selective by taking advantage of surface waves. We also report the discovery of the enhancement by several orders of magnitude of the energy density close to an interface at a particular frequency as well as the enhancement of the radiative flux between two interfaces when surface phonon polaritons can be excited. These results lead to the design of a novel class of infrared incandescent sources with potential applications in spectroscopy and thermophotovoltaic energy conversion.

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R É S U M É

Il est généralement admis que le rayonnement thermique est spatialement et temporellement incohérent. Nous montrons ici qu'en présence d'ondes de surface, il faut remettre en cause cette idée. Il est possible de concevoir des sources incandescentes qui sont directionnelles et spectralement sélectives. Nous décrivons également la découverte de l'exaltation de plusieurs ordres de grandeur de la densité d'énergie près d'une interface à une fréquence particulière ainsi que l'exaltation du flux radiatif échangé entre deux surfaces lorsque des ondes de surface existent. Ces résultats permettent d'envisager une nouvelle génération de sources incandescentes avec des applications possibles à la spectroscopie et à la conversion d'énergie par effet thermophotovoltaïque.

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1. Introduction

Blackbody radiation is a topic that has been extensively studied at the beginning of the twentieth century [1,2]. It played an important role in the development of quantum physics with the pioneering work of Planck [1]. It also had many applications with the development of lighting with incandescent light sources. After these developments, a number of features have been attributed to thermal sources: light produced by thermal sources has been considered to be both

E-mail address: jean-jacques.greffet@institutoptique.fr.

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spatially and temporally incoherent. Energy transfer mediated by radiation, i.e. radiative heat transfer, is bounded by the value of the blackbody flux density given by σT^4 where σ is the Stefan–Boltzmann constant. In the last twenty years, with the advent of nanophotonics, this old topic has been revisited and a new vision has emerged. It has been recognised that incandescent sources could produce narrow beams and that the emitted spectrum could be tailored at will. In other words, it has been recognised that thermal sources can be spatially and temporally partially coherent, particularly when surface waves such as surface plasmons or surface phonon polaritons can be excited. It has also been discovered that the presence of surface waves modifies drastically the blackbody radiation in the near field, namely, at a distance from the source smaller than $\lambda/2\pi$. The energy density can be increased by several orders of magnitude and becomes quasimonochromatic for some materials at distances in the order of 100 nm. As a consequence, the radiative heat flux between two surfaces can be increased by orders of magnitude as the gap distance enters the nanoscale regime. All these effects pave the way to novel applications such as the design of thermal sources with unprecedented new properties or thermophotovoltaic electricity generation. These properties can be analysed in the framework of fluctuational electrodynamics that was introduced by Rytov to discuss thermally generated radiowaves [3,4]. An account of recent work in the infrared range can be found in refs. [5–8].

2. Spatially coherent thermal radiation

The title of this section would have been very provocative twenty years ago. Thermal radiation was taken as the typical example of incoherent radiation. Obviously, blackbody radiation in a cavity much larger than the wavelength is incoherent. However, thermal light emitted by a surface can become partially spatially and temporally coherent. Before summarising how this unexpected property was discovered, let us briefly summarise why thermal light is expected to be incoherent. If we consider the light emitted by an interface separating an absorbing medium from vacuum, it is known that the emitted light can be characterised by a specific intensity or radiance. This is given by the product of the blackbody (equilibrium) radiance, which depends only on the material temperature and the emissivity of the material. The emissivity of a plane interface is given by $1 - R$, where R is the intensity Fresnel reflection factor. Since the emissivity is smaller than 1, the emitted radiance is always smaller than the blackbody radiance. Since the emissivity is a smoothly varying function of angle and frequency, the emission is broadband and quasi isotropic. According to the Wiener–Kinchine theorem, which establishes that the power spectral density is the Fourier transform of the time correlation function, a broad spectrum means that the time correlation is very short and therefore that the field is incoherent. Conversely, a laser with a very narrow spectrum produces light with a long coherence time. Similarly, a laser can be very directional, while thermal radiation is quasi isotropic. This is also related by a Fourier transform to the spatial correlation function of the fields.

The above discussion based on the concept of emissivity is based on a purely phenomenological description. It is possible to gain further insight by analysing thermal radiation with an antenna point of view: electromagnetic fields are generated by time-dependent currents. With this point of view, the source of the thermally emitted electromagnetic fields is the current density due to the thermal random motion of charges (ions, electrons) in the material. This is the origin of the thermal radiation in an electrodynamic picture. This approach was already introduced by Lorentz in 1906 [2], but there was no correct statistical description of the current fluctuations available at that time. In order to actually compute the thermal fields, the random thermal currents need to be known. They are given by the fluctuation–dissipation theorem, which yields the following form for the correlation of the current density j [9]:

$$\langle j_n(\mathbf{r}, \omega) j_m(\mathbf{r}', \omega') \rangle = 2\pi \delta(\omega + \omega') \delta(\mathbf{r} - \mathbf{r}') \delta_{nm} 2\omega \epsilon_0 \text{Im}[\epsilon(\mathbf{r}', \omega)] \Theta[T, \omega] \quad (1)$$

where the brackets denote the ensemble average, ϵ is the permittivity, $\Theta[T, \omega] = \hbar\omega / [\exp(\hbar\omega/k_B T) - 1]$ and T is the temperature. This type of modelling of the thermal radiation was introduced by Rytov in order to analyse thermal radiation in radioelectricity [3]. The same formalism has been used by Lifshitz to study Casimir forces by real metals [10]. Note that the procedure that we have just outlined is a Langevin model. Indeed, computing the fluctuating electromagnetic fields by including in Maxwell equations a random source is similar to the introduction of a random force in Newton's equations to model the Brownian motion. It is interesting to note that the random currents are spatially delta-correlated. This is a consequence of the assumption of locality (no spatial dispersion) of the optical response. Hence, it seems reasonable to assume that close to the source, the radiated fields are spatially incoherent. This is the usual assumption in the statistical optics textbooks [11,12]. As a consequence, the fields emitted by different points of the source cannot interfere in the far field so that the thermal sources are expected to be quasi isotropic. This assumption is actually not valid in some cases, as we will see below.

2.1. Questioning Kirchhoff law validity

The investigations of coherence of thermal emission were triggered by the work of D. Maystre published in 1976. He predicted [13] the phenomenon of total absorption by a metallic sinusoidal grating. It was predicted that a grating ruled on a silver or a gold surface can absorb all the light incident at a specific angle for a particular frequency. This phenomenon was very surprising as a gold sinusoidal surface illuminated at 633 nm with a maximum slope of a few percents has a reflectivity that drops from over 0.97 (it is a mirror) out of resonance to less than 0.01 on resonance. Yet, it was confirmed

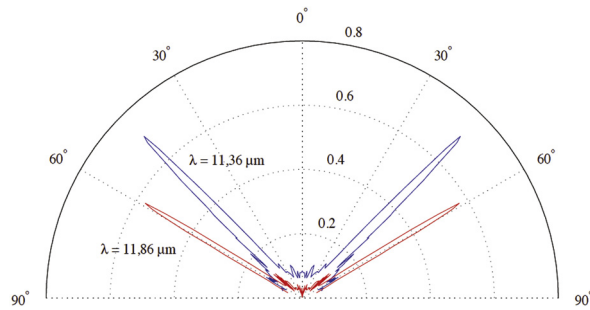


Fig. 1. Angular emission pattern of a SiC grating at two different frequencies. It is seen that the emission pattern is similar to the emission pattern of an antenna with narrow lobes. Reproduced from [21].

experimentally by Hutley and Maystre [14]. The angular width of the absorption dip is on the order of 1 degree. This absorption is due to the resonant excitation of a surface wave that exists in the near field of the surface. The grating is used to couple efficiently a propagating plane wave with a surface wave. When I became aware of this experiment, I used to teach thermal radiation and I had a background of optics and solid-state physics. Hence, I realised that if Kirchhoff's law was valid, by heating such a surface, one should observe thermal emission in a very well-defined direction at a well-defined frequency. In other words, thermal radiation should be partially coherent both spatially and temporally. The other possibility was that directional emission would not be observed so that Kirchhoff's law would not be valid for such resonant surfaces. The validity of Kirchhoff's law is an old topic that belongs to the category of questions asked by students and professors teaching basics rather than by professors attending hot-topic conferences. The original paper by Kirchhoff [15] provides a rigorous but lengthy derivation that uses the Helmholtz reciprocity principle. Yet, there was no rigorous proof of the Helmholtz reciprocity principle in the framework of radiometry. Many textbooks [16,17,9] derivations of Kirchhoff law are simplified and have a flaw, as discussed by M. Huetz-Aubert [18]. In summary, either Kirchhoff's law was not valid, at least for this type of resonant surface, or one had to admit that thermal sources could be coherent.

2.2. Thermal emission by SiC gratings

While this was an interesting academic question, it did not become an active research topic until I met M. Olivier, who was interested in finding surfaces with highly anisotropic radiative properties. He first proposed to investigate the optical properties of SiC gratings. By analogy with total absorption due to surface plasmons, he expected to observe total absorption in the mid-infrared at a particular angle due to surface phonon polaritons. The first results did not show the expected total absorption angular peak [19], so he contacted me to help analysing their data. It turned out that the grating had been designed using a perturbative model that was not accurate in the resonance regime. Further numerical analysis explained the data and led to the design of a new grating with total absorption, reproducing Maystre's results by exciting a surface phonon polariton instead of a surface plasmon polariton [20]. This grating was studied both in reflectivity and in emissivity and the dip observed in reflectivity produced as expected an angular peak in the emission pattern, thereby providing the first evidence of spatially coherent thermal emission [20]. As the first data were limited in angular resolution, F. Marquier built a new setup during his PhD work. Fig. 1 reproduces his experimental measurement of the angular pattern of the emission performed [21,22]. It is seen that the emission of this hot surface is characterised by lobes that are reminiscent of the angular lobes of radiowave antennas.

The experimental result was consistent with Kirchhoff's law. Hence, it was interesting to try to derive a general proof. From previous works, it was sufficient to prove the Helmholtz reciprocity principle [18] in the framework of radiometry. Since reciprocity is a theorem in electrodynamics of continuous media for media with symmetric permittivity tensors, the question is whether it is possible to export this property from electrodynamics to radiometry. It turns out that the link between radiometry and electrodynamics had been established using statistical optics [23,24], thereby establishing the foundations of radiometry. It can be shown that the radiance, which depends on both the propagation direction and the spatial position, is the Wigner transform of the fields. By using this connection between radiometry and electrodynamics, it was possible to prove the validity of Kirchhoff's law for any interface separating two media satisfying reciprocity [25]. We later became aware of another general proof of Kirchhoff's law, previously derived by Rytov with a different approach [3].

2.3. Origin of the spatial coherence

While Kirchhoff law predicts an angular emission pattern with a narrow angular peak, it does not explain what is the underlying physical mechanism producing coherence. In particular, one may be tempted to think that the grating is responsible for the induced coherence. In order to gain some physical insight, we used with R. Carminati [26] the fluctuational electrodynamics framework that has been briefly outlined in the introduction: the fluctuating electromagnetic fields are generated by the random motion of charges in local thermodynamic equilibrium at temperature T . In the case of SiC, the fields emitted in the infrared are due to the random motion of the Si and C atoms that carry a partial charge. This random

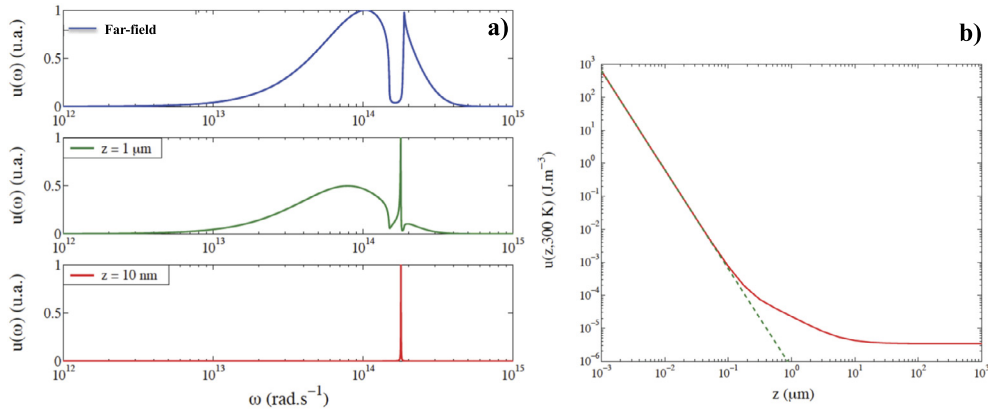


Fig. 2. Spectral energy density above a SiC–vacuum interface. a) Normalised spectra of the energy density at three distances. Top: far field, middle: 1 μm , bottom: 10 nm. b) Energy density integrated over all frequencies as a function of the distance to the interface z (logarithmic scale). Reproduced from [28].

motion produces a distribution of random electric dipole moments that excite surface phonon polaritons (the analog of surface plasmons). The key to understand the origin of the spatial coherence over large distances is to realise that surface phonon polaritons are modes localised along the surface. Hence, a localised random source (e.g., a single dipole) excites a mode that propagates along the interface. The electromagnetic field of a given mode exists at different points P and M of the surface and therefore induces a long-range correlation of the fields at these points. This simple argument suggests that the coherence length is the decay length of the surface phonon polariton along the surface. It also stresses that the coherence is due to the surface wave, not to the grating. The periodic grating only serves the purpose of revealing the spatial coherence of the fields close to the surface. It is merely a coupler just like a waveguide coupler diffracting the mode of a waveguide into a plane wave.

This discussion is correct when dealing with the field at a distance in the order of $\lambda/2\pi$, which is the typical size of the height of a grating. Let us stress that the correlation length strongly depends on the distance z to the interface. For distances larger than $\lambda/2\pi$, the correlation length is in the order of $\lambda/2$ as in vacuum, as one would expect. By contrast, in the near-field region with $d \ll \lambda/2\pi$, the correlation length becomes smaller than $\lambda/2$.

3. Blackbody radiation in the near field. Temporal coherence

A second unexpected property is the existence of a spectral peak in the density of energy of equilibrium radiation very close to a surface supporting a surface phonon polariton. This unexpected feature was discovered by K. Joulain by exploring the spectrum of the energy density in the near field of a SiC/vacuum interface [27] in collaboration with A. Shchegrov. Again, using the electrodynamics model, the fields can be computed at any distance from the interface. It has been known for many years that the electromagnetic fields thermally emitted can be larger than the blackbody radiation in the near field [3] of the sources. The physical mechanism is simple: the energy density close to a dipolar emitter is due to the large electromagnetic fields decaying as $1/r^3$ in the near-field regime $r \ll \lambda/2\pi$. Since any material volume element much smaller than the wavelength can be modelled by a random dipole moment, this argument is quite general. The results are shown for a SiC–vacuum interface on Fig. 2. The novelty is to observe: i) a significant modification of the spectrum with the distance z to the interface, ii) a dramatic enhancement of the energy density at the peak of the surface phonon polariton. We plot in Fig. 2a the spectra normalised by their peak value. It is seen that the spectrum becomes quasimonochromatic. That particular frequency is the frequency of the asymptote of the surface wave dispersion relation. From the Wiener–Kinchine theorem, a narrow spectral peak means that the electromagnetic field is partially temporally coherent. The coherence time, which is proportional to the inverse of the width of the peak, is essentially the decay time of the surface wave.

We now consider the energy density integrated over the spectrum. The result is shown in Fig. 2b. It is seen that the energy density increases by several orders of magnitude. From statistical physics, it is known that the energy density is the product of the density of states and the mean energy of a harmonic oscillator. Hence, our result means that the local density of electromagnetic states increases locally at a particular frequency. This is due to the existence of a very large number of additional electromagnetic modes as compared to vacuum: the surface waves. Of course, as surface waves are bounded to the interface, this increase in energy density depends on the distance to the interface z . To analyse these effects, we were led to revisit the concept of density of electromagnetic states in regions surrounded by lossy media [5,29].

The existence of strong fields thermally excited close to the surface has been observed experimentally in the group of Y. De Wilde. The experiment uses a metallic tip brought close to the surface. The tip scatters the near-field and send it to a Cassegrain objective. By scanning the tip, it is possible to image the near-field distribution emitted by the sample [30]. This experiment is very challenging as the typical power is on the order of a few pWs and one essentially works with an AFM in tapping mode with a sample at 250 $^{\circ}\text{C}$. The group was able to overcome the difficulties and produced images of the local

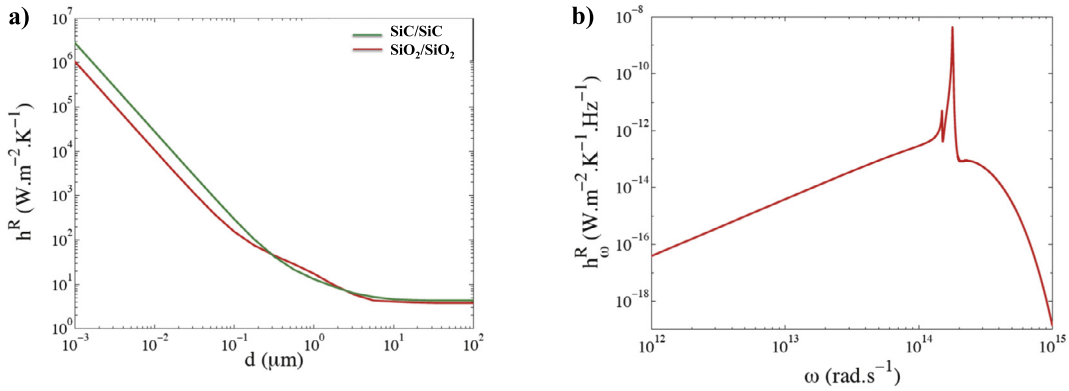


Fig. 3. Radiative heat transfer between two plane surfaces separated by a distance d . a) Heat transfer coefficient as a function of the gap width d . It is seen that the flux varies as $1/d^2$ in the small distance regime. Reproduced from [28]. b) Spectrum of the heat transfer coefficient.

density of electromagnetic states on a gold stripe. The spectral narrowing in the near field has been verified experimentally recently by two groups [31,32].

4. Enhanced radiative heat transfer

4.1. Role of surface waves in the radiative heat flux at nanoscale

A direct consequence of the enhancement of the energy density is the possibility of increasing the heat transfer beyond the usual far-field value. A simple situation is to bring a subwavelength sphere at temperature T_1 close to an interface separating vacuum from a medium at temperature $T_2 > T_1$. The small sphere will be exposed to electric field and magnetic fields that are orders of magnitude larger than in a blackbody at the same temperature. Hence, it will be heated more efficiently. We studied [33] this situation and predicted fluxes enhanced by several orders of magnitude when approaching the surface. Actually, it was already known that the heat flux exchanged between two metallic surfaces at temperatures T_1 and T_2 could exceed the flux $\sigma(T_1^4 - T_2^4)$ when the distance d is smaller than the wavelength. Experiments had been reported [34,35]. A complete theory [36] had been developed by Polder and van Hove. This theory predicts an asymptotic behaviour of the flux at short distance proportional to $1/d^2$. It is very similar to the Lifshitz model [10] of Casimir force, which is a momentum flux between two surfaces. There were no quantitative comparison with theory apart from an inconclusive measurement [37]. However, only the heat transfer between metals had been considered. Furthermore, the large increase in the flux in the very short-distance regime below 50 nm had not been observed.

Our key finding was the discovery of the role of surface waves. We found that the heat transfer between dielectrics supporting surface phonon polaritons is much larger than between metals [28,38]. While metals can support surface plasmons, their energy is in the near ultraviolet, so that they cannot be thermally excited. The contribution of the surface phonon polariton led to an enhancement of one order of magnitude of the flux [28,38]. We show in Fig. 3 the heat transfer coefficient $h^R = \phi/(T_2 - T_1)$, which is the flux divided by the temperature difference assuming $T_2 - T_1 \ll T_1$. It is clearly seen that the flux can be increased by orders of magnitude as compared to the far-field case. The second unusual feature of this enhanced radiative flux is its spectral content. As expected from what has been seen in the density of energy, most of the energy is transferred to a narrow spectral range corresponding to the surface phonon polariton resonance. As a follow up of this study, we investigated the Casimir force between two metals. Indeed, the formalism to compute the energy flux or the flux of the z -component of the momentum (i.e. the force) between two interfaces is very similar. This study showed that the Casimir force in the short distance regime is dominated by the contribution of surface plasmons to the density of states [39].

The discovery of the role of surface phonon polaritons in the radiative heat transfer at the nanoscale triggered a new generation of experiments using dielectrics supporting surface phonon polaritons instead of metals. The first data obtained in the group of G. Chen at MIT and by E. Rousseau in our group in collaboration with J. Chevrier's group were taken with silica and showed very good agreement with the Polder–van Hove theory [40,41]. The field continues to grow, and recent results have been reported with different materials and different geometries [42–44].

4.2. Physical analysis

The asymptotic form of the flux predicts a variation proportional to $1/d^2$ when the gap width goes to zero. Its validity has been questioned [45]. Here, we aim at giving a simple explanation of the origin of this asymptotic behaviour. To this aim, we use the framework introduced by Landauer and Buttiker to deal with ballistic electronic transport [46]. In short, each plane wave characterised by a wavevector (k_x, k_y) and a frequency ω can be viewed as a mode that contributes to the flux between two thermostats at temperature $T_1 = T + \Delta T/2$ and $T_2 = T - \Delta T/2$. If $\Delta T \ll T$, it can be shown that the

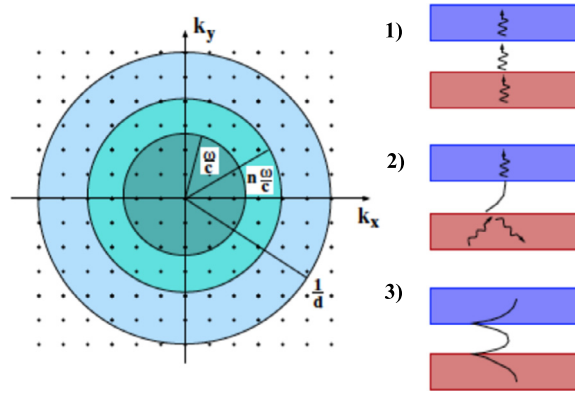


Fig. 4. Origin of the near field enhancement. The flux is proportional to the number of modes. For $k_{\parallel} < \omega/c$, the modes are the propagating waves depicted in inset 1. For $\omega/c < k_{\parallel} < n\omega/c$, the modes correspond to the frustrated total internal reflection as depicted in inset 2. For $k_{\parallel} > n\omega/c$, the modes are pure evanescent modes, as depicted in inset 3.

flux Φ due to a single mode is given by $\Phi = g\Delta T$, where g is the quantum of thermal conductance given by $g = \frac{\pi^2 k_B^2 T}{3h}$ where k_B is Boltzmann’s constant and h is Planck’s constant. Coming back to the case of two parallel surfaces at ambient temperature, the flux is given by $Ng\Delta T$, where N is the number of modes. With this point of view, we can revisit the Stefan–Boltzmann constant σ appearing in the flux between two black surfaces with area S , $\Phi = S\sigma(T_1^4 - T_2^4) \approx S4\sigma T^3 \Delta T$. It can be checked that $4S\sigma T^3 = Ng$, where $N = \frac{2\pi S}{\lambda_T^2}$ is the number of modes. Here, λ_T is the Planck wavelength given by $hc/k_B T$. It is seen that the number of modes is roughly given by the ratio S/λ_T^2 .

We now discuss what changes in the near field. Electromagnetic modes with wavevectors $\mathbf{k}_{\parallel} = (k_x, k_y)$ such that $k_x^2 + k_y^2 > \omega^2/c^2$ have a pure imaginary wavevector component $k_z = (\omega^2/c^2 - k_{\parallel}^2)^{1/2}$ and cannot propagate over distances larger than $1/|k_z|$. Indeed, the transmission factor through the gap decays as $\exp(-k_{\parallel}d)$ for large k_{\parallel} , so that $1/d$ appears as a cutoff. If $k_{\parallel}d < 1$, the energy can tunnel through the gap. This is illustrated in Fig. 4 where we represent the plane (k_x, k_y) . For small gaps, evanescent modes with $k_{\parallel} < 1/d$ can contribute to the flux by tunnelling. It is clearly seen in the figure that their number increases as the area of the disk with radius $1/d$. A crude estimate of the increase of the flux can be done when moving from far field to a gap distance of 100 nm. The cutoff moves from $1/\lambda$ to $1/d$. Taking $\lambda = 10 \mu\text{m}$ and $d = 100 \text{ nm}$, we expect a four orders of magnitude enhancement of the flux. This is an upper bound corresponding to the case of a transmission factor unity for all frequencies. Achieving this type of performance is currently an active research topic.

The study of radiative heat transfer at the nanoscale raises another fundamental question. What is the difference between conduction and radiation? As the gap distance goes to zero, one expects to recover the conduction regime. So where is the limit between the two regimes? Is there a fundamental difference between the two phenomena? To explore this question, we performed a molecular dynamics simulation studying the flux between two SiO₂ nanoparticles [47]. This is a polar crystal so that we included a direct Coulomb interaction between nuclei. While we expected some discontinuous transition between a conduction regime and a radiative regime, we found that there is a continuous transition between far-field radiation and conduction at contact due to the near-field interaction. In other words, the traditional difference between conduction and radiation vanishes at the nanoscale in the case of polar materials. Both are a manifestation of the Coulomb interaction between nuclei. The situation is different in the case of non-polar crystals where the Coulomb force is shielded. In that case, it has been predicted that there may be (acoustic) phonon tunnelling. This is currently an active research topic [48].

5. Outlook

In this brief overview, a few key properties of radiation at the nanoscale have been selected and simple physical pictures have been put forward. We have stressed the key role played by surface waves. The physics of radiative heat transfer at nanoscale is now well understood and the field is now driven by applications and rapidly expanding. One of the directions of current research is the design of a new generation of infrared (IR) incandescent emitters. When looking for infrared sources, there are either expensive coherent sources such as quantum cascade lasers and optical parametric oscillators or incandescent sources such as hot plates and globars. The properties discussed in this paper pave the way to the design of cheap and compact IR sources with unprecedented properties such as directionality [49,50], controlled spectrum [51], improved efficiency [52,53] and high-frequency modulation [54,55]. While the brightness of these sources is limited as the number of photons per mode is limited by Bose–Einstein distribution, the emitted power is quite significant. For instance, a source at 1000 K with emissivity 1 and an emitting area of 1 cm² emits 43 mW/sr in the spectral range [9, 10] μm .

Another application is in the field of energy conversion. Thermophotovoltaics is a conversion energy scheme using photovoltaic cells with low bandgaps illuminated by incandescent sources. Its major advantage is the possibility to tune the

emission spectrum of the sources in order to match the gap of the cell. It is thus possible to avoid losing energy due to either photon excess energy or the impossibility to absorb photons with energies smaller than the cell gap. Thermophotovoltaics can be used to extract energy from low-temperature sources so it can be an alternative to thermoelectricity. In this context, the remarkable radiative properties of nanostructures could be very useful [56,57].

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