Degree of polarization of the thermal near field generated by arrays of metallic nanoparticles

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By employing a rigorous multiple-scattering Green's tensor formalism, we present exact calculations of the degree of polarization of the blackbody radiation in the presence of one or more spheres whose dimensions are smaller or comparable to the radiation wavelength. For a single metallic sphere the thermal field is partially polarized close to the surface due to near-field effects and becomes totally unpolarized away from the surface. Interestingly, for a finite chain of metallic spheres, the degree of polarization of the thermal field decreases compared to the case of a single sphere. A measurement of the degree of polarization by means of thermal-radiation scanning-tunnelling microscopy can provide an image of a heated surface with subwavelength resolution.

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I. INTRODUCTION

Ever since Glauber established[1] the quantum theory of optical coherence, the stochastic properties of the electromagnetic (EM) field have received a great deal of attention (see Ref. 2 and references therein). In the work of Glauber, complete coherence is achieved only if the EM field is totally polarized. Recently, Wolf[3,4] introduced a unified theory of partial coherence and polarization of the EM field in which case complete coherence is possible for totally unpolarized fields. According to this theory, the state of coherence and polarization of a stochastic light beam changes on propagation as a result of the correlations in the emission of radiation within the source. However, Wolf's definition of coherence changes upon application of local-linear deterministic transformations and alternative definitions for the degree of coherence should be used instead.[5,6]

A typical example of stochastic EM field is the thermal (blackbody) radiation which results from the random thermal motion of charges which generates random current sources inside a material.[7] Blackbody radiation is traditionally considered as an unpolarized and incoherent type of radiation. However, the recent advances in micro- and nanofabrication have made possible the realization of artificial periodic structures emitting thermal radiation, which, in the far field, is characterized by a high degree of coherence and narrow emission lobes (large directionality in space).[8] The most intriguing phenomena related with the coherence and polarization of the thermal field occur at distances very close to material surfaces where the EM near field is dominant.[9–11] Such phenomena are the dependence of the thermal-emission spectrum on the distance from the surface,[12] spatial-correlation lengths which can be much shorter or much larger than wavelength,[13] and deviations of the spatial coherence of thermal light from standard coherence theory.[14]

A less explored phenomenon is the polarization of the thermal-radiation near field. Although thermal radiation from a surface of homogeneous material is unpolarized in the far field, periodically patterned surfaces may emit partially polarized thermal radiation.[15] In the near field, the degree of polarization is greatly enhanced above the surface of a material supporting surface waves such as surface plasmons or polaritons.[16] In this work, we study the degree of polarization of thermal radiation emitted from *finite* material bodies (such as spheres) since the assumption of *infinite* bodies (such as planar surfaces) leads to unnatural situations such as the existence of polarized thermal radiation at infinite distances from the body.[16] In particular, we find that the degree of polarization close to a metallic nanosphere assumes nontrivial (nonzero) values around the surface-plasmon (SP) frequency of the sphere. When more spheres are placed together so as to form a linear one-dimensional (1D) chain, we find that the thermal near field becomes *less* polarized and coherent. Finally, as an application, we show that the experimental measurement of the degree of coherence provides an image of the emitting surface with subwavelength resolution.

II. THEORY

Our approach is based on the framework of fluctuational electrodynamics[17] and more specifically, on the fluctuation-dissipation theorem for the EM field.[7,18] The latter relates the cross-spectral correlation function (tensor) $W_{ii'}(\mathbf{r},\mathbf{r}';\omega) = (1/2\pi)\langle E_i(\mathbf{r},\omega)E_{i'}(\mathbf{r}',\omega)\rangle$ of the fluctuating electric field E_i at thermal equilibrium with the Green's tensor of the system $G_{ii'}$, i.e.,

$$W_{ii'}(\mathbf{r},\mathbf{r}';\omega) = \mu_0 \hbar \omega^2 \coth\left(\frac{\hbar\omega}{2k_B T}\right) \mathrm{Im}\, G^{EE}_{ii'}(\mathbf{r},\mathbf{r}';\omega), \quad (1)$$

where $i,i'=x,y,z$. ω is the angular frequency, T the temperature, μ_0 is the magnetic permeability of vacuum, $\hbar$ the reduced Planck's constant, k_B the Boltzmann's constant, and $G^{EE}_{ii'}(\mathbf{r},\mathbf{r}';\omega)$ is the component of the full Green's tensor $G_{ii'}$, which provides the electric field at $\mathbf{r}$ due to an electric dipole source at $\mathbf{r}'$. The Green's tensor for a given collection of N scatterers is calculated on the basis of a rigorous EM multiple-scattering theory and explicit relations can be found elsewhere.[19,20] We note that the correlation tensor $W_{ii'}$ given by Eq. (1) presumes thermal equilibrium between the collection of particles and the surrounding medium.[7] This means that apart from the blackbody radiation emitted by the particles, $W_{ii'}$ also includes the radiation which is emitted by the host medium and subsequently (multiply) scattered off by the particles.

In order to study the polarization of the thermal near field, we choose the definition of the three-dimensional (3D) degree of polarization $P(\mathbf{r})$ at a point $\mathbf{r}$ in space;[21] the two-dimensional (2D) definition of Wolf[4] is suitable for propagating beam-like stochastic fields and cannot account for near-field effects in which case all three orthogonal components of the electric field are appreciable. Namely, the 3D degree of polarization is given by[16,21]

$$P^2(\mathbf{r};\omega) = \frac{3}{2}\left\{\frac{\mathrm{Tr}[\mathbf{W}^2(\mathbf{r},\mathbf{r};\omega)]}{\mathrm{Tr}^2[\mathbf{W}(\mathbf{r},\mathbf{r};\omega)]} - \frac{1}{3}\right\}. \quad (2)$$

According to the above definition of the 3D degree of polarization, an EM field can be considered as polarized when the electric-field vector lies within a plane but not necessarily along a specific direction in space.[21,22] This property is not met in 2D definitions of P.[23] However, for beam-like and transverse EM fields there is a one-to-one correspondence between the 3D and 2D degrees of polarization and are therefore both valid measurements of the (intensity weighted) average correlation between the field components at a single point.[24]

The corresponding degree of coherence between two points $\mathbf{r}$ and $\mathbf{r}'$ in space is taken to be[6]

$$C^2(\mathbf{r},\mathbf{r}';\omega) = \frac{\mathrm{Tr}[\mathbf{W}(\mathbf{r},\mathbf{r}';\omega)\mathbf{W}(\mathbf{r}',\mathbf{r};\omega)]}{\mathrm{Tr}\ \mathbf{W}(\mathbf{r},\mathbf{r};\omega)\mathrm{Tr}\ \mathbf{W}(\mathbf{r}',\mathbf{r}';\omega)}. \quad (3)$$

We note that for scalar fields, the degree of coherence $C(\mathbf{r},\mathbf{r}')$ between two points $\mathbf{r}$ and $\mathbf{r}'$ is a quantitative measure of the ability of these points to produce visible interference fringes of the field intensity in Young's two-pinhole experiment.[4] However, for the EM field which is, in general, a vector field, the degree of coherence is a measure of the modulation of the four Stokes parameters. This implies that although intensity fringes may not be visible in Young's experiment, the occurrence of polarization modulation, i.e., modulation in at least one of the Stokes parameters, results in a nonzero degree of coherence.[6]

III. RESULTS AND DISCUSSION

We begin our study by considering a metallic sphere described by a local, Drude-type dielectric function

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}. \quad (4)$$

A sphere of this type supports SP excitations at frequencies $\tilde{\omega}_l \simeq \omega_p\sqrt{l/(2l+1)}$, $l=1,2,\cdots$ which dominate the absorption and scattering spectra of collections of such particles. The 3D degree of polarization $P(\mathbf{r};\omega)$ as defined in Eq. (2) is depicted in Fig. 1. It is evident that for a wide range of frequencies, the thermal near field is unpolarized except in the regions around the dipolar ($l=1$) SP resonance which is located at $\tilde{\omega}_1=\omega_p/\sqrt{3}=0.577\omega_p$. Within this region, the degree of polarization is appreciable even for large distances ($r\sim 2c/\omega_p$) from the surface of the sphere. The existence of the SP modes leads to a collective motion of the charges/currents at the surface of the sphere and, therefore, the emitted electric-field fluctuations are primarily radially polarized with respect to the sphere giving rise to a partially polarized thermal field. We note that similar effects are expected for ionic and semiconductor spheres supporting phonon-polariton and exciton-polariton surface waves, respectively.

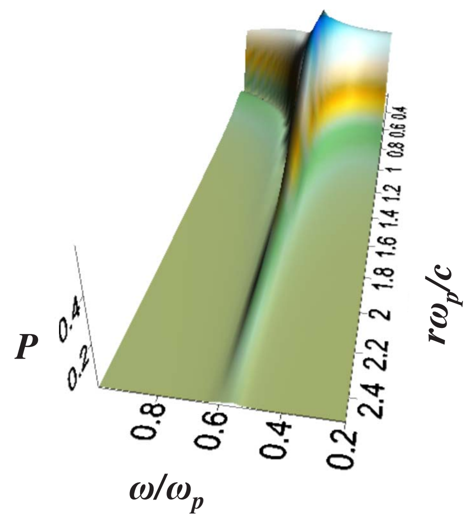

FIG. 1. (Color online) Degree of polarization P of the thermal field emitted a Drude-type sphere ($S=0.2c/\omega_p$) as a function of normalized frequency ω/ω_p and distance $r\omega_p/c$ from the center of the sphere.

When many thermal emitters (metallic spheres) are placed in periodic fashion so as to form a 1D grating or a chain, the emission of thermal radiation becomes highly directive due to its large degree of spatial coherence.[8,14] In order to explore the degree of polarization of the thermal-radiation field in such a system, we consider a linear, periodic array of metallic spheres (chain) with radius $S=0.2c/\omega_p$ and lattice constant (period) $a=c/\omega_p$ (see Fig. 2). Figure 3 shows the degree of polarization P for the dipolar SP frequency ($\omega/\omega_p=1/\sqrt{3}$) as a function of the distance r from the center of the middle sphere of the chain (see Fig. 2), for different chain lengths (N is the number of the spheres in the array). It is evident the degree of polarization assumes, more or less, the same value on top of the surface of a single sphere regardless of the number of spheres in the chain suggesting a much

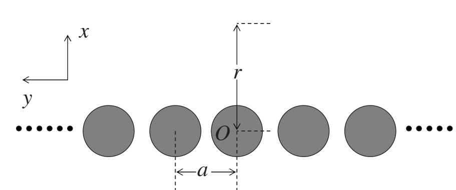

FIG. 2. A linear finite array of metallic spheres arranged periodically with period a. The degree of polarization P is calculated along a line being normal to the array and passing through the center of the middle sphere (x axis).

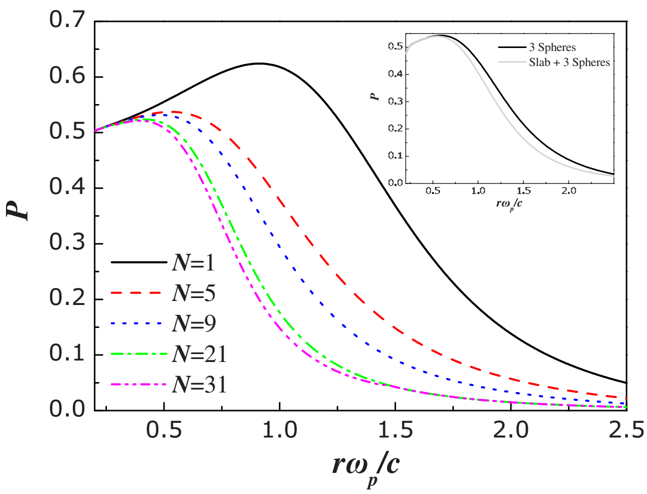

FIG. 3. (Color online) Degree of polarization $P(r)$ for frequency $\omega/\omega_p=0.57$ as a function of the distance from the center of the middle sphere, for linear arrays of spheres with $S=0.2c/\omega_p$ (those depicted in Fig. 2) of various sizes (number of spheres N). The inset shows $P(r)$ for three metallic spheres on top of a quartz ($\epsilon=2.1$) substrate (thick line). The thin line refers to the case of three spheres without substrate.

weaker contribution of the thermal fluctuations coming from the rest of the spheres of the chain. The value of P at the surface of the sphere depends on the radius of the sphere and for very large spheres approaches 1/4 which is the value of P at a planar surface.[16] We also observe that the thermal field becomes less polarized as the chain length increases. Figure 4 shows the degree of spatial coherence for the same chains of particles as in Fig. 3.

It is clear that C is greatly enhanced due to SP excitation and varies with distance, more or less, the same way as P. We note that, in both Figs. 3 and 4, as the number of spheres N increases, P and C decrease and the respective curves approach the corresponding curve of the infinite chain (the curves for $N=21$ and $N=31$ are practically the same).

This can be understood as follows. If the array is viewed as a single thermal source, its statistical properties are modified by the interaction of the EM field fluctuations of each

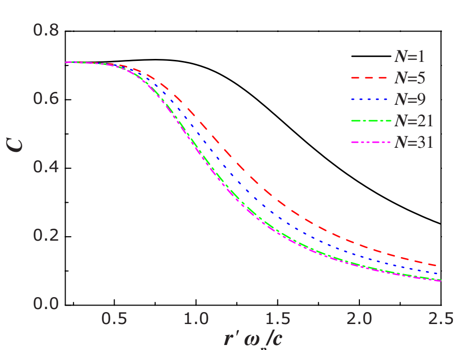

FIG. 4. (Color online) Degree of coherence $C(r,r')$ for frequency $\omega/\omega_p=0.57$ and $r=0.2c/\omega_p$ from the center of the sphere, as a function of the normalized distance $r'\omega_p/c$ from the center of the middle sphere, for linear arrays of spheres with $S=0.2c/\omega_p$ (those depicted in Fig. 2) of various sizes (number of spheres N).

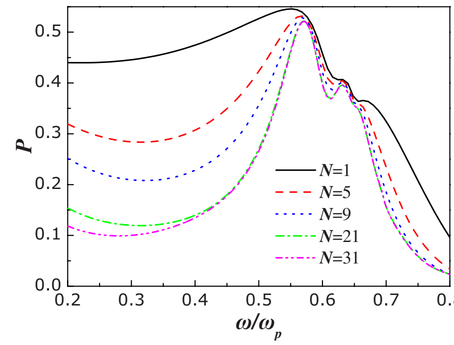

FIG. 5. (Color online) Spectrum for the degree of polarization P for linear arrays of spheres with $S=0.2c/\omega_p$ (those depicted in Fig. 2) of various sizes (number of spheres N). P is calculated at a distance $r=0.4c/\omega_p$ from the center of the middle sphere for each array.

sphere with its neighboring ones; this interaction leads to a broader absorption/emission bandwidth around the SP region which is translated as a larger value of the variance of the current fluctuations in the array as a whole. The latter results in the progressive decrease in the spatial coherence toward a saturated value as the chain length increases. In other words, as the chain length increases, the thermal field in the middle of the chain becomes more "isotropic" and, hence, less polarized and coherent. We note that the decrease in P very close to the sphere surface is due to the quasistatic contribution to the correlation function which is larger than that of the SPs.[9,16]

In the inset of Fig. 3 we also show P for the case where the spheres are in touch with a quartz ($\epsilon=2.1$) substrate (it is compared against the case of spheres without a substrate). It is evident that the presence of a substrate has little influence on the functional form of $P(r)$ apart from an overall decrease, which is due to the infinite extent of the substrate, i.e., the presence of a substrate has more or less the same effect as an increase in the chain length.

In order to understand the decrease in P with the chain length (Fig. 3), we have plotted the spectrum of the degree of polarization, $P(\omega)$, for a specific distance $r=0.4c/\omega_p$ from the center of the middle sphere and for various chain lengths (see Fig. 5). The three distinct peaks in all spectra of Fig. 5 correspond to the dipole (lower frequency), quadrupole, and octapole (higher frequency) SP resonances. It is evident that all the thermal-radiation field curves for $N>1$ are below the corresponding curve for $N=1$. The underlying mechanism for the above results is illustrated as follows. For a single sphere and at a particular point in space, the radial component of the thermal EM fluctuations (the W_{xx} component in our case) dominates over the tangential ones (the W_{yy} and W_{zz} components in our case) resulting in the calculated degree of polarization. When many spheres are brought together to form a chain, the radial EM fluctuations stemming from the other (more distant) spheres contribute mostly tangential than radial components (defined relative to the same coordinate system used for a single sphere) at a particular point in space. This leads to an overall decrease in the degree of polarization. The degree of polarization P continues to decrease the more spheres are added to the chain until it

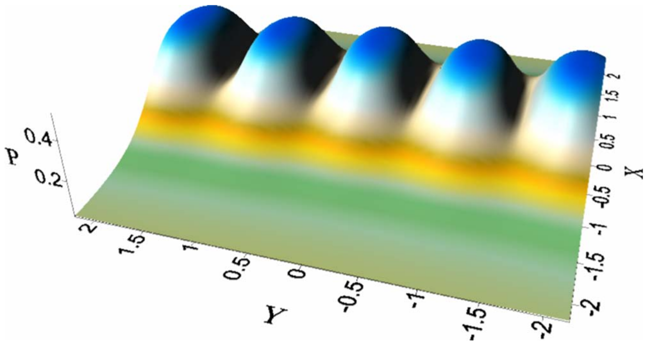

FIG. 6. (Color online) Degree of polarization P calculated within the xy plane at a distance $z=0.5c/\omega_p$ above a linear array of metallic spheres, for frequency $\omega/\omega_p=0.57$. The coordinates x and y are in c/ω_p units.

finally converges to a specific value in the limit of infinite chain length.

Next, in Fig. 6, we calculate the degree of polarization P for the SP frequency, within a xy plane at a distance $z=0.5c/\omega_p$ above a linear array of 11 metallic spheres. It is evident that the variation in P depicts an image of the array: the maxima of P correspond to the sphere centers while the minima to the midway points between the spheres. This picture of the degree of polarization can be potentially useful in polarization-sensitive near-field imaging by making use of thermal-radiation scanning tunnelling microscopy (TRSTM)[25] which measures the thermal-radiation near field above a heated sample. Namely, by rotating the sample so that the microscope tip be oriented along each of the three orthogonal directions (x, y, and z) one can measure, each time, a different component of the thermal electric-field fluctuations (we remember that a conical tip chiefly measures the electric-field along the tip axis) and from these measurements deduce the degree of polarization. The main advantage of TRSTM over traditional near-field scanning optical microscopy lies in the lack of external illumination in TRSTM. An image of the degree of polarization is free of topographic artifacts stemming from the dissimilar temperatures of the sample and the microscope tip (as it is the case when measuring directly the thermal-radiation field[25]) since the degree of polarization does not depend on the temperature.

IV. CONCLUSION

In conclusion, based on fluctuational electrodynamics and a rigorous multiple-scattering Green's-tensor technique, we have shown that the blackbody near field emitted by a sphere supporting surface waves, such as a metallic sphere supporting SPs, is partially polarized. In the case of a linear chain of metallic spheres, the blackbody field becomes less polarized and coherent as more spheres are added to the chain. The degree of coherence can provide an image of the underlying emitting structure with subwavelength resolution within the spectral regime of the surface-wave resonances.

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