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Procedures for the measurement of the extinction cross section of one particle using a Gaussian beam

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ABSTRACT

Two procedures for the measurement of the extinction cross section (ECS) of one particle using a slightly focused Gaussian beam have been introduced and numerically tested. While the first one relies on previously introduced ideas and has close connection with the optical theorem, the second procedure is new and is mostly related with light measurements where the detector collects much of the energy of the incident beam.

Both procedures prove to be valid and somehow complementary up to particle sizes of the order of the beam waist, thus enlarging the capability of simple measurement set-ups based on Gaussian beams for the estimation of the ECS of one particle.

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1. Introduction

The calculation and measurement of the ECS of one single particle is a common problem in light scattering theory [1,2]. The generalization of the definition of ECS to the case of non-plane incident wave is also a topic widely addressed. Of course, one of the cases of major interest is for Gaussian beams. Without going very back in time, some relevant references are (in chronological order) [3–9]. A complete review of the topic is done in [10].

Nowadays, the ease of use of Gaussian beams focused on one particle suggests that it would be useful to devise approximate experimental procedures for the measurement of the ECS of the particle based on that configuration. In our work we will consider a linearly polarized Gaussian beam slightly focused (divergence angles up to a maximum of 10°) as the incident excitation.

In this context, we propose two approximate methods for the measurement of the ECS of a single particle by

means of the detection of light without and with the particle placed on the focus of the incoming Gaussian beam. One of the procedures was already introduced in Ref. [5] and relies on the light detection only in a small angle in the forward direction. Conversely, the second procedure that we will propose will be more adequate when light collecting angles are wider.

In Ref. [5], the validity of several approximate expressions for the calculation of the ECS by using Gaussian beams is discussed in great detail, both from an analytical and from a numerical point of view. Restricting ourselves to numerical tests, the present work checks the accuracy of the two methods we will propose by means of computer calculations and analyzes how they compare with analytical results for spherical particles using Mie theory. The work is based on the explicit numerical calculation of the Poynting vector of the waves reaching the light detector. This detector is considered to be of finite aperture, subtending a well-known angle from the center of the Gaussian beam, just the precise position where the particle is placed. For this purpose, specific and precise numerical methods have been developed. The necessary procedures

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for the detailed development of our ideas are presented as follows:

2. Development of specific numerical methods to handle the problem

For the calculation of the Poynting vector at any point in space, the interference between the beam illuminating the particle and the subsequently scattered field must be explicitly formulated.

In our thought experiment (Gedankenexperiment) the illumination is performed with a Gaussian beam with small divergence angle $\sin(\delta_0) = NA_{\text{eff}}$, whose mathematical expression (within the standard paraxial theory) is:

$$E(z, \rho) = E_0 \frac{\omega_0}{\omega(z)} \exp\left(\frac{-\rho^2}{\omega^2(z)}\right) \exp i\left(\frac{k\rho^2}{2R(z)} - \Phi_G(z)\right) \exp i(kz), \quad (1)$$

where ω_0 is the beam radius in the focal plane, $R(z) = z\left(1 + \left(\frac{z_R}{z}\right)^2\right)$, $z_R = \frac{k\omega_0^2}{2}$, $\Phi_G(z) = \tan^{-1}\left(\frac{z}{z_R}\right)$, ρ is the radial distance from the axis and k the wave-vector.

The previous formulae assume the Z axis to be beam axis and the X and Y directions to be in the transverse directions. We will assume that the Gaussian beam is linearly polarized, with the electric field having always the direction of the constant unit vector \hat{e}_x . Within this approximation, the corresponding magnetic field is:

$$H = \frac{\hat{e}_z \times E}{c\mu_0}, \quad (2)$$

with c the speed of light and μ_0 the magnetic permeability of vacuum. Given the wavelength λ , power P_0 and $NA_{\text{eff}} = \sin(\delta_0)$ the peak amplitude E_0 can easily be found by using

$$\omega_0 = \frac{\lambda}{\pi\delta_0} \quad (3)$$

and

$$P_0 = \frac{\pi E_0^2 \omega_0^2}{4c\mu_0} \quad (4)$$

We plan to use the Mie theory for the calculation of the scattered field, but this theory is developed for a spherical particles excited by a linearly polarized plane wave (of amplitude E_p). Generalized Mie theories [10] can tackle the situation of spatially inhomogeneous illumination. Yet, if the particle can be considered as homogeneously polarized, the use of Mie theory is well justified [2]. In any case, since our illumination is not a plane wave, we have to estimate E_p for the particle, when it is being excited by the Gaussian beam.

When the particle (with radius a) is centered in the beam waist, even when the particle is small, taking $E_p = E_0$ is not the best choice since, as the Gaussian profile has a maximum on axis, this assumption always overestimates the value for E_p . We propose calculate E_p as follows.

a. Find the power incident on a centered circle of radius a ,

$$P_a = P_0 \left(1 - \exp\left(\frac{-2a^2}{\omega_0^2}\right)\right) \quad (5)$$

b. Find the constant ('mean') value for the electric field E_m that corresponds to the flux of the power P_a across the area πa^2 ; this is

$$E_m = \sqrt{\frac{2P_a \eta_0}{\pi a^2}}, \quad \text{with } \eta_0 = 377 \Omega. \quad (6)$$

c. Assume $E_p = E_m$.

Besides, this approach does not require the size of the particle to be very small. Once E_p is estimated, the Mie theory can be used, giving for the scattered field

$$E_s(r, \theta, \phi) \approx E_p \frac{\exp i(kr)}{-ikr} X(\theta, \phi). \quad (7)$$

The Mie theory provides the calculation of the angular term, the vector scattering amplitude $X(\theta, \phi)$, by assuming a spherical shape for the particle, with radius a . This topic is very well known; in Ref. [2] all important details can be found. Particularly, in our work we will make continuous use of the result regarding to the number of terms (in the final series development) that is enough for an accurate representation of the scattered field [11]. This result relates explicitly the number of terms to the radius of the particle (more terms required as the particle gets bigger). Thus, we consider our calculations of the scattered field as 'exact' since we have fulfilled the requirements imposed by the condition discussed in Ref. [11].

According to our previous choice for the axis, the scattered field will have the three spatial components but, clearly the longitudinal one (Z) will be negligible in the far field with respect to the other two. Thus, finally, when there is a particle present in the path of the Gaussian beam, we will consider the Jones vector of the total field reaching the light detector in the far zone to be

$$\begin{pmatrix} E_G + E_s|_x \\ E_s|_y \end{pmatrix}, \quad (8)$$

where E_s is the scattered field generated by the Gaussian beam E_G .

This formulation shows that, since the Cartesian components of the scattered field (in the far field) are being calculated, we can calculate the interferences (sum) between this scattered field and the incident Gaussian beam. The scenario is like in Figure 3.7 of Ref. [2], with the difference of assuming illumination by means of a Gaussian beam, not with a plane wave like there (see Fig. 1).

A careful analysis of Fig. 1 illustrates that the computation of the Poynting vector of the resulting field on the points of the sphere and subsequent integration over the area defined by the aperture of the detector, allows us to calculate the power collected by this detector. According to Fig. 1, the collecting area will be defined on the imaginary

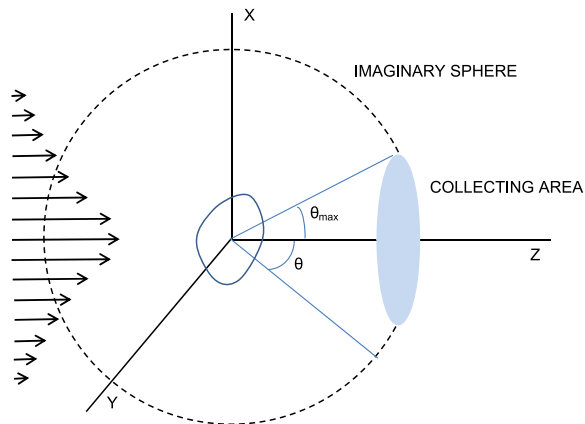


Fig. 1. Geometry associated to our Gedankenexperiment. The sphere is in the far field and we will assume that the detector collects all the energy propagating up to a maximum (limiting) angle θ_{\max} .

sphere by the conditions

$$0 \leq \theta \leq \theta_{\max}, \quad 0 \leq \phi < 2\pi. \quad (9)$$

A set of scripts and functions was written in MATLAB language, implementing the above ideas.

As an illustrative example, by using the cited programs, we have computed the power reaching the detector with or without the particle at the focus. Fig. 2(a) plots as a continuous red line the watts collected up to the angle θ_{\max} indicated by the abscissa, when the particle is at the center of the waist. The wavelength is $1 \mu\text{m}$ (Au complex refractive index $0.26 + 6.97i$). The incident power is $P_0 = 10.0 \text{ mW}$, with a beam waist radius of $\omega_0 = 1.58 \mu\text{m}$ (beam divergence angle $\delta_0 = 11.5^\circ$, i.e. 0.2 rad). The radius of the particle is $a = 0.3 \mu\text{m}$ and the radiation due to the particle is calculated by the Mie theory (under the assumptions previously discussed) by estimating the amplitude of the plane wave as indicated in the steps a–b–c previously. Similarly, up to the angle θ_{\max} indicated by the abscissa, the dotted blue line shows the amount of light collected without the particle (thus, for the incident beam alone). The continuous green line corresponds to the corresponding amount of scattered light. The same results are sketched in Fig. 2(b) using a double logarithmic scale to better illustrate the behavior for small collecting angles.

Q3 It is worth considering in detail the meaning of the results represented in this Figure, since they will be a clear indication of the overall accuracy of our calculations. Following the notation used in Ref. [12], the Poynting vector reaching any point of the detector area, corresponding to the incident field alone (no particle present) is written as S_i . The integral of this Poynting vector up to the angle θ_{\max} indicated by the abscissa is the dotted blue line in the Figures. Similarly, when the particle is placed in the beam waist, the Poynting vector of the resulting wave is written $S = S_i + S_s + S_{\text{ext}}$, where S_s stands for the scattered field and S_{ext} is the term that arises because of interaction between the incident and scattered waves. The integral of this Poynting vector S up to the angle θ_{\max} indicated by the abscissa is the red line in the Figures, while the green line is the integral for S_s . Thus, the role of the interference term

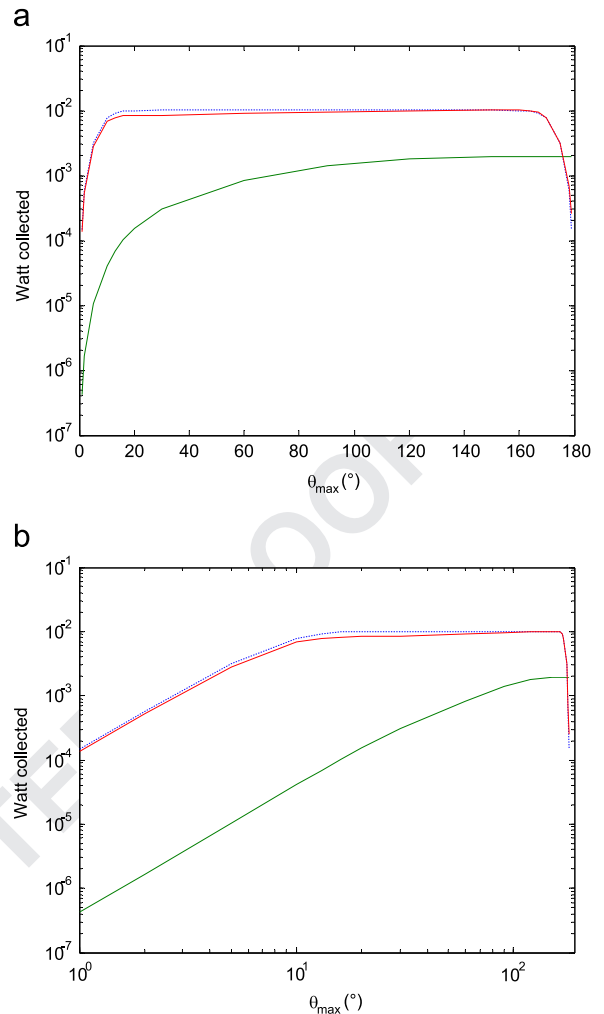


Fig. 2.

S_{ext} is evidenced, since the red line cannot be obtained simply by adding the green and the blue ones.

There are several interesting features of the graph:

- One can see the increase of the values for the incident beam as the collecting angle increases; the increase is noticeable up to values of θ_{\max} of the order of δ_0 ; then, strictly speaking, the values still increase up to 90° ; for θ_{\max} above 90° , the decrease has the opposite tendency than from 0° to 90° .
- One can see that the scattered field increases monotonically as the collecting angle increases, as expected.
- One can check that the final value of the scattered power (i.e. for 180°) coincides with the value predicted by the Mie Theory for our amplitude of the plane wave E_p and our particle radius.

In summary, we are confident that we have the numerical tools for the precise calculation of the fields scattered by one particle as well as for the accurate

1 integration of the resulting Poynting vectors on integration
3 areas defined by angular zones centered on the focus of
the incoming Gaussian beam.

3. Statement of the two procedures

7 Assume we have a configuration like in Fig. 1. When the
9 particle is not present, we have simply the incident beam
and we name the collected power P_i^c . With the particle at
11 the center of the waist of spot size (radius) ω_0 , the power
at the detector will be P_t^c . Note that we use a superscript in
13 our formulas since they are related to the light effectively
‘collected’ by the detector, as defined by its position
15 and size.

17 As indicated in the introduction, the first procedure we
propose here was already devised in Ref. [5]. In this
Reference the justification of the method is also presented;
19 there is no precise indication of the size of the light
detector: it should be as small as possible since the pro-
21 cedure is based on measuring the intensity strictly in the
forward direction. In summary, by measuring the power
23 received by a detector placed in the forward direction with
respect to the incoming Gaussian beam, according to what
25 we call ‘Procedure I’, the estimated value for the extinction
cross section is:

$$C_{\text{ext}}^I = \pi\omega_0^2 \left(1 - \frac{P_t^c}{P_i^c}\right) = \pi\omega_0^2 \left(\frac{P_i^c - P_t^c}{P_i^c}\right) \quad (10)$$

31 With our numerical methods, we will be able to study
the influence of the size of the detector, which is equiva-
33 lent to say to its angular size (θ_{max} in our expressions). This
influence is important in practice because there is a com-
promise between the size of the detector and the amount
35 of collected light, since if we want to restrict ourselves
only to the forward direction, we have to tend to a very
37 small detector, leading to small intensity measurements.
Thus, our numerical procedures can be used to evaluate
39 quantitatively this compromise, as will be shown below.

41 The second procedure we will study in the present
work is, to the best of our knowledge, a new proposal. It is
43 based in a few simple ideas (to be immediately exposed)
and constitutes an alternative approach to the problem.
45 We will see that this second procedure is more adequate
than the first when the collecting angle increases, allowing
47 more light detection. Thus, it constitutes an alternative
approach that will be analyzed and compared with the
49 first procedure by means of our numerical methods.

51 Let us establish precisely this ‘Procedure II’. The main
underlying idea behind the method is one elementary
53 result from the theory of Gaussian beams: the fraction of
the power of a Gaussian beam of waist size ω_0 , incident
over a centered circle of radius r is

$$\exp\left(\frac{-2r^2}{\omega_0^2}\right) \quad (11)$$

57 Thus, following the mathematical notation just intro-
59 duced, our assumption in our method is: the presence of
the particle of unknown effective radius r at the center of
the beam waist of size ω_0 reduces the power at the

detector from the value P_i^c to the value P_t^c . This is equiva- 63
lent to write

$$\frac{P_t^c}{P_i^c} = \exp\left(\frac{-2r^2}{\omega_0^2}\right) \quad (12) \quad 65$$

67 from where

$$r^2 = -\frac{\omega_0^2}{2} \ln\left(\frac{P_t^c}{P_i^c}\right) = \frac{\omega_0^2}{2} \ln\left(\frac{P_i^c}{P_t^c}\right) \quad (13) \quad 69$$

71 and finally

$$C_{\text{ext}}^{II} = \pi r^2, \text{ which is } C_{\text{ext}}^{II} = \pi \frac{\omega_0^2}{2} \ln\left(\frac{P_i^c}{P_t^c}\right) \quad (14) \quad 73$$

75 Note that we are finding an ‘effective’ radius r as
77 defined by the fact that it is the one that would give to our
beam the extinction we are measuring.

79 Besides, assuming weakly radiating particles P_i^c/P_t^c is
close to 1 and by taking only the first term of the series
81 expansion of the ‘ln’ in expression (14), we can easily
obtain an ‘approximate formula’ for Procedure II:

$$C_{\text{ext}}^{II'} \approx \pi\omega_0^2 \left(\frac{P_i^c - P_t^c}{2P_t^c}\right) \quad (15) \quad 83$$

85 As before, for Procedure I, our computations will allow
87 checking the adequacy of these different proposals, with-
out restriction for different sizes and particle materials.

89 In summary, we are proposing in the present work the
comparison between three formulas, corresponding to two
91 different conceptual approaches. Procedure I leads to
expression (10) and Procedure II gives expression (14),
93 which can be approximated by formula (15). Thus, the
difference between Procedure I and the ‘approximate for-
95 mula’ for Procedure II is contained in the two factors

$$\frac{P_i^c - P_t^c}{P_i^c} \text{ and } \frac{P_i^c - P_t^c}{2P_t^c} \quad (16) \quad 97$$

99 that multiply the common term $\pi\omega_0^2$. In the following,
our numerical techniques will be used to evaluate quan-
101 titatively the suitability of any of the three choices:
expression (10), expression (14) and expression (15).

4. Test and comparisons for spherical particles

105 Fig. 3 shows the results of using the two procedures for
107 estimating the cross section of a 50 nm gold particle in the
range of wavelengths from 200 to 1000 nm. The optical
109 constants of Au are taken from Ref. [13]. For such particle
and spectral range, the extinction cross section is domi-
111 nated by Au interband transitions below 500 nm and a
dipolar plasmon resonance around 520 nm. We consider a
113 Gaussian beam with $\delta_0 = 5.7^\circ = 0.1$ rad. It is important to
note that, as we are assuming $\delta_0 = 5.7^\circ$ to be constant, the
115 size of the waist increases linearly with the wavelength,
since $\omega_0 = \frac{\lambda}{\pi\delta_0}$ for Gaussian beams. For comparison, we will
117 assume light detectors collecting up to five different
angular widths, so that the angle θ_{max} can be 1° , 2° , 6° , 10°
119 or 15° wide as seen from the center of the beam waist.
Thus, for the smaller collecting angles only part of the
121 incident beam will be effectively collected when no par-
123 ticle is present in the beam path, as indicated in previous

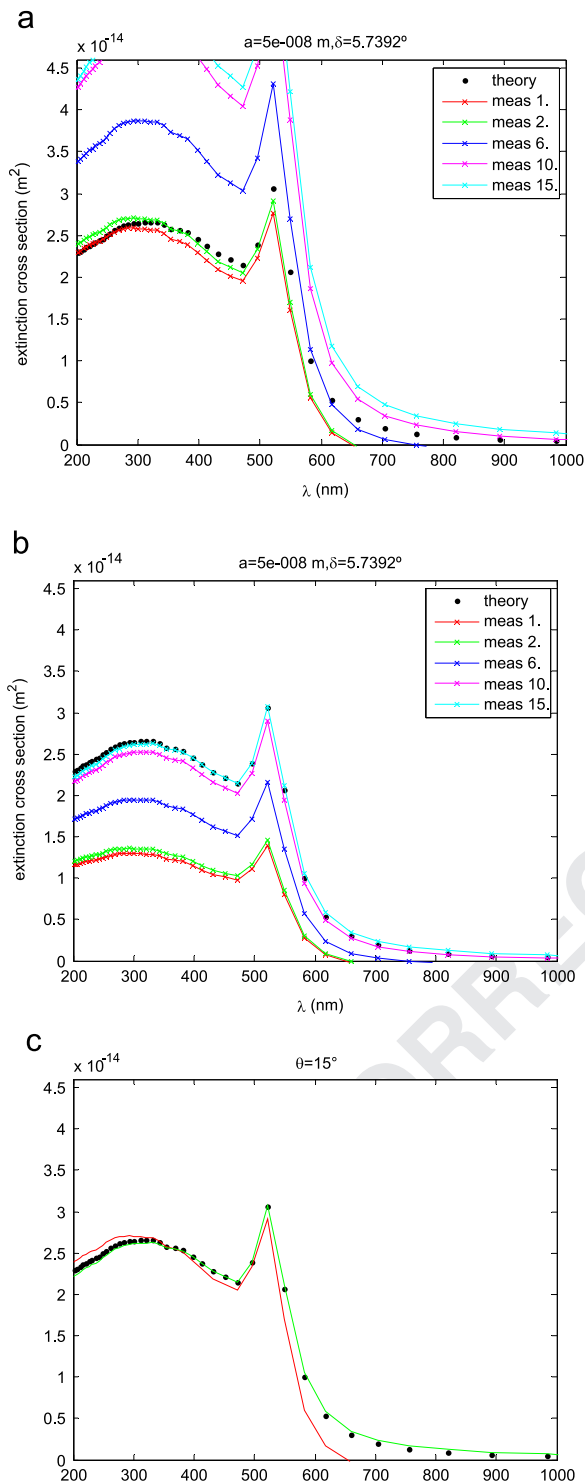


Fig. 3. (a) Results of using Procedure I for a 50 nm radius gold particle, depending on the light collecting angle. (b) Results of using Procedure II for a 50 nm radius gold particle, depending on the light collecting angle. (c) Results of Procedure I for 2° (red line) compared with those of Procedure II for 15° (green line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 2. Fig. 3(a) shows the results of using Procedure I: the actual results given by the Mie theory are presented as black dots and the values with Procedure I using the energies up to $1^\circ, \dots, 15^\circ$ as continuous lines. Fig. 3(b) shows the same kind of results for Procedure II. It is clear that results for Procedure I are accurate for small angles and those of Procedure II for big angles, as could be expected from the very definition of them. For better comparison between the results, Fig. 3(c) shows the results of Procedure I for 2° (red line) with those of Procedure II for 15° (green line). The values obtained for the 'approximate formula' for Procedure II (formula 15) are virtually the same as the exact ones (formula 14) and only one green line corresponding to formula (14) is shown.

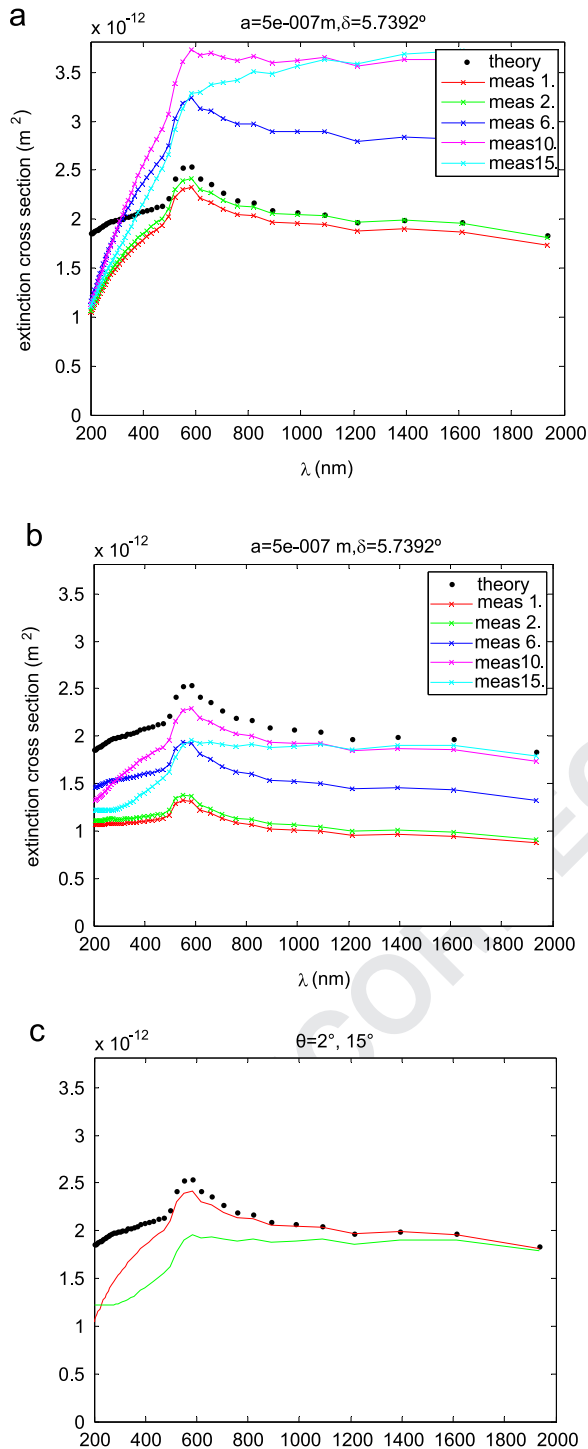
Keeping the particle size of 50 nm, the results show exactly the same kind of behavior for water (refractive index around 1.33) and for Si (refractive index around 3.5) and therefore the plots are not shown for brevity.

Fig. 4 shows the results for the estimation of the cross section of a 500 nm gold particle in the range of wavelengths from 200 to 2000 nm using the same type of Gaussian beam.

Now the behavior of the two Procedures is more complex. One reason is evident and expected: when the size of the particle is of the same order (or bigger) than the waist of the beam, the accuracy is poor; this is what we see for wavelengths below 600 nm. Under these conditions, it cannot be assumed that the particle is polarized homogeneously and replacing the Gaussian beam by a plane wave is not well justified. Above 600 nm the results of Procedure I are accurate for small detector angles, as in the previous case. However, now the results for Procedure II do not become more accurate if the detection angle is arbitrarily increased. On the contrary, it seems there is some optimum collection angle where the results of this Procedure present the best accuracy (somewhere around 10°). The results for the 'approximate formula' of Procedure II (expression 15) differ from the exact ones (those of expression 14) for short wavelengths; since they tend to be even less accurate, the results given by (15) will not be shown in the plots.

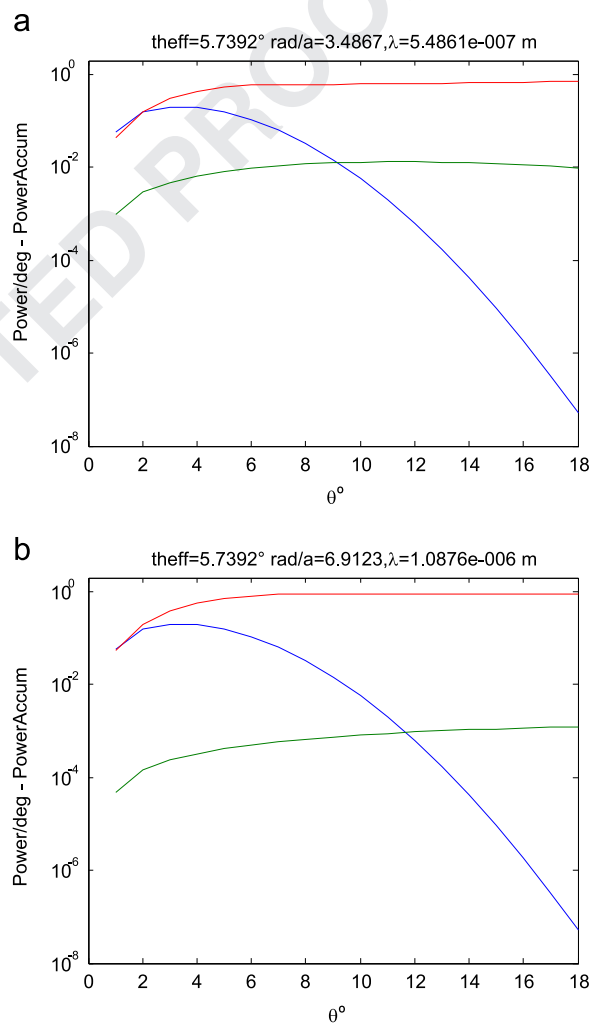
To find an explanation for the worse performance of this Procedure II, Fig. 5(a) plots together two kinds of light power collected, for 548 nm wavelength. For the incident beam (blue line) and scattered beam (green line) the computed 'power collected per angular degree' is shown while the red line corresponds to the 'power accumulated' corresponding to the resulting total field (incident plus scattered). We call 'power collected per angular degree' (for some angle θ expressed in degrees) the light power comprised between $\theta - 1^\circ$ and θ° (for $0 \leq \phi < 2\pi$). We call 'power accumulated' (for some angle θ expressed in degrees) the light power comprised between 0° and θ° (again for $0 \leq \phi < 2\pi$). In Fig. 5, the θ values in the X axis go from 1° to 18° . One can see that the power related to the incident beam (blue line) is equal to the power related to the scattered beam (green) at 9° approx. The light being collected above this angle is primarily light scattered by the particle and therefore cannot be interpreted as a

1 contribution to the shadow casted to the Gaussian beam.
 2 Thus the best estimation of the ECS at this wavelength
 3 using Procedure II is obtained if the collection angle is



57 **Fig. 4.** (a) Results of using Procedure I for a 500 nm radius gold particle,
 58 depending on the light collecting angle. (b) Results of using Procedure II for
 59 a 500 nm radius gold particle, depending on the light collecting angle. (c)
 60 Results of Procedure I for 2° (red line) compared with those of Procedure II
 61 for 15° (green line). (For interpretation of the references to color in this
 figure legend, the reader is referred to the web version of this article.)

62 limited to avoid scattering contributions that do not
 63 remove energy from the incident beam (see Eq. (12)). Fig. 5
 64 (b) plots the same quantities for the wavelength of
 65 1087 nm. Now the equality of powers takes place at about
 66 12° . However, a good estimation of the ECS is obtained for
 67 both 10 and 15 deg. Now, the scattered light collected
 68 when the detection angle is increased above this critical
 69 angle represents only a small contribution (compare the
 70 orders of magnitude) to the already collected power. In
 71 this situation of relatively small scattering at large detec-
 72 tion angles, results from Procedure II are insensitive to the
 73 maximum detection angle because P_t^c does not remarkably
 74 change above certain angle. Thus, Procedure II will be
 75 accurate for any detection angle provided the detector
 76 covers most of the region where the incident and scattered
 77 fields significantly interfere.



81 **Fig. 5.** (a) For 548 nm. Power collected per angular degree of the incident
 82 beam (blue line) and of the scattered beam (green line). Power accumu-
 83 lated corresponding to the resulting field (red line). (b) For 1087 nm.
 84 Power collected per angular degree of the incident beam (blue line) and
 85 of the scattered beam (green line). Power accumulated corresponding to
 86 the resulting field (red line). (For interpretation of the references to color
 87 in this figure legend, the reader is referred to the web version of this
 88 article.)

Again, for this new particle size of 500 nm we have computed the same kind of results for water, for Si (and even for Ag). The behavior is completely similar to what we have shown for Au and we do not present the plots.

5. Conclusions

We have numerically tested two procedures for the estimation of the extinction cross section of one particle using a slightly focused Gaussian beam. Both procedures require placing the particle in the center of the waist and measuring the decrease of the light reaching the detector (assumed centered on the path of the beam). If the two power measurements (without and with the particle on the light path) are P_i^c and P_t^c and the beam waist radius is ω_0 , the two procedures give for the extinction cross section the expressions (10) and (14) (respectively for Procedures I and II). With the approximation introduced in (15), the difference between Procedures I and II is virtually contained in the formulas (16).

We have presented explicit comparisons for gold spheres of radius of 50 and 500 nm, in the range between 200 and 2000 nm, for an incident beam with $\delta_0 = 5.7^\circ$ and collecting angles of 1° , 2° , 6° , 10° and 15° . We have also performed series of calculations for spheres of H_2O , Si and Ag, for different beam divergence angles, obtaining in all the cases the same conclusions regardless the properties of the material.

The analysis of these results allows us to summarize the conclusions as follows.

- 1) We have verified the validity of both procedures to determine the ECS of particles with sizes significantly smaller than the beam waist. Besides, we have been able to illustrate the role of the angle of the light detector in the measurements.
- 2) We have shown that Procedure I is accurate when the angle subtended by the light detector from the center of the waist is very small (say about 2°) while Procedure II is accurate when the light detector is such that collects all the light of the incident beam up to the angle where its intensity is similar to that of the scattered. Procedure II gives also accurate results for larger collecting angles provided the light scattered at these angles does not significantly modify P_i^c .

- 3) Since we have a quantitative assessment of the validity of the two approaches, we can use these ideas to evaluate the precision related to one particular practical configuration.
- 4) Our study introduces the possibility of using Procedure II when the intensity of the light detected (associated to the small collecting angle of Procedure I) is a limiting practical issue.

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