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Development of an in situ ITER dust diagnostic based on extinction spectrometry: Dedicated light scattering models

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ABSTRACT

The quantity of dust produced in fusion devices must be characterized in order to address their consequences on the operation of the machine as well as on safety issues. This paper presents the work currently in progress to develop light scattering models of dust as well as a light extinction spectrometer to monitor dust concentration and size distribution in the vacuum vessel.

1. Introduction

During tokamak operation dusts production needs to be characterized and monitored from the operation point of view (fuel recovery, maintenance) as well as for safety issues (off-normal events: air ingress, hydrogen and carbon explosion hazards) [1–3]. In current tokamaks, dust properties studies (composition, shape, size distribution, etc.) rely on recovery techniques (manual or robotized) and outline physical–chemical analyses. In the frame of the ITER program where dust mass inventory limits have been defined [4], there is a need to develop online techniques to monitor dust quantity on the hot surfaces and more generally in the vacuum vessel [1,3]. Development of ITER relevant optical particle characterization (OPC) techniques, during plasma shutdowns, is nevertheless not a trivial task due to (i) limited and difficult optical accesses, large distances, vibrations, magnetic field strength; (ii) expected complexity and variety of dusts shapes: homogeneous and layered spheres (from plasma facing component sublimation and vapor condensation), cauliflower shapes and flakes (from the breaking of the codeposited layers), fibres (from carbon–fibre–carbon plasma component material) or nanotubes [1,2]; (iii) the lack of data on dust composition and the optical properties of mixed materials (in ITER, dusts are foreseen to be made of W, Be, and C with the presence of metallic impurities (Fe, Cr, etc.) and some oxides (WeO, AlO_3, etc.)); (iv) large size distribution range expected for dusts (with diameter $D = 10$ nm–10 µm), presence of large flakes (10 µm to few millimetres) which should nevertheless rapidly deposit in case of an air ingress for instance.

In this paper we report the work in progress to develop an 'Extinction Spectrometry' technique to characterize dusts in suspension in ITER vacuum vessel. Section 2 presents the numerical tools to predict the basic light scattering properties of dust (cross-sections, scattering and polarization diagrams) and which are required by all OPC techniques (ellipsometry, diffractometry, extinction spectrometry, etc.). Section 3 details the principle of the extinction spectrometry technique as well as the data inversion procedure we have developed to recover dust size distribution and concentration.

2. Optical and light scattering properties of dusts

Prior to the development of any optical diagnostic of dust concentration and size distribution it is necessary to first model their optical properties (complex refractive spectra) and then, their light scattering properties (electromagnetic calculations).

2.1. Complex refractive index

The determination of dusts complex refractive index $\tilde{m}$ is a crucial task as this parameter has a determinant influence on their scattering properties and it is an input parameter of the light scattering models. Most OPC techniques require knowing in advance the particle refractive index for at least one wavelength (over a band spectrum for the extinction spectrometry). The problem is that there is very few data available on the refractive index of dusts

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encountered in tokamaks. Our first strategy is then to collect in the literature the refractive index of the different elements composing dusts; then, it is to use effective medium theories to estimate the dust effective refractive index for different compositions; finally, to infer the refractive index from extinction measurements.

The refractive index spectra of carbonaceous, Beryllium, Tungsten particles, and their oxides, are totally different each other [2]. The optical properties of the two latter materials (Be and W) are also rather different each other in the visible and the near-infrared region (NIR), but they are similar in the middle-infrared region (MIR) [2]. From these spectra we can already infer that the scattering properties of dust must be highly dependant on their relative composition in carbon and metallic elements (which reinforce the interest to determine dust composition with laser-induced breakdown spectrometry [3]).

Dusts may be composed of different materials. This can be managed, in some extend, by the scattering models (i.e., aggregates of spheres of different materials). Materials mixtures can also be modeled by effective medium theories like the Maxwell–Garnett or the Bruggeman ones [5]. These theories treat the problem of an inhomogenous medium as a two-components mixture composed of inclusions embedded in an homogeneous matrix. Inclusions have the same dielectric permittivity but may be different in volume, shape and orientation. Our simulations with this approach [2] have shown that, when considering a carbonaceous particle with an increasing volume fraction of spherical inclusions of tungsten, up to 20–30% of tungsten in volume, the medium refractive index spectra remains somewhat similar to the carbonaceous one (whatever the mean values drastically increase). For higher concentrations in tungsten the spectra are totally different from the carbonaceous and the tungsten ones [2].

Another way to determine the refractive index spectra of dusts is to perform experimental analyses. Unfortunately, the determination of the refractive index of powders is also an extremely delicate task (they will be activated and tritiated). This requires at least obtaining some dusts samples. When it will be the case, the procedure is to measure the spectral hemispheric-reflectivity of dusts (ideally from UV to MIDIR) and to use Kramers–Kronig integral to reconstruct the complex refractive index spectra [5].

An alternative solution will be to use the extinction spectroscopy technique itself to recover dust refractive index [6] (i.e., inversion with an iterative procedure on refractive index [7]). This latter solution requires also a dust sample and the measurement with a reference technique [6] of the dust size distribution.

2.2. Light scattering theories

Optical diagnoses of dust size distribution and concentration require an accurate modeling of their scattering properties. We need to describe the scattering properties of homogeneous, stratified or heterogeneous spheres, spheroids, fibres, flakes like shapes, fractal or compact aggregates of elementary spheres [1,2], see Fig. 1.

The Lorenz-Mie Theory (LMT) which solves the basic problem of the scattering of a plane electromagnetic waves by a spherical homogeneous particles has been extended during the two last decade to model the scattering of an arbitrary beam by a spherical homogeneous sphere [8,9], a multilayered or heterogeneous sphere [10,11], a spheroid [12], etc. This theory uses a separation variable method to solve Maxwell's equations with the appropriate boundary conditions. The predictions of this theory (internal fields, radiation pressure...) are widely and intensively used in all academic and technical fields as a reference or as a tool of practical interest to model light scattering of droplets, bubbles, industrial beads, fibres, etc.

To simulate the light scattering properties of dusts and flakes encountered in tokamaks the use of a spherical particle model

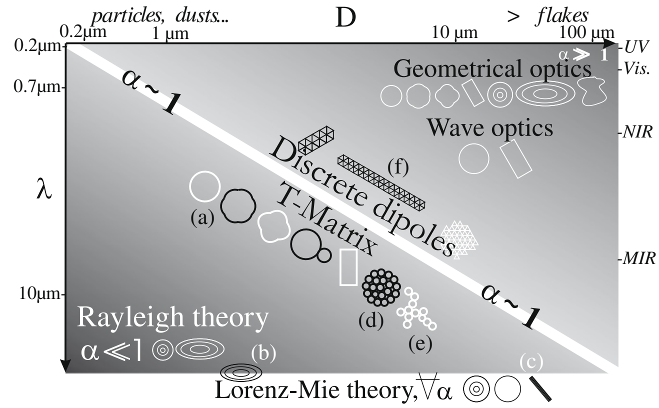

Fig. 1. Validity chart of various light scattering theories regarding to the particle size parameters and shapes: (a) spheres, (b) spheroids, (c) fibres, (d) compact aggregates (cauliflower shapes), (e) dilute aggregates (chains, fractals), (f) cubic or platelets forms (flakes).

may be considered as questionable. Perfectly spherical particles are known to generate powerful surface waves and morphological dependence resonances (SW-MDR, cavity modes) that are never observed for irregularly shaped particles. Nevertheless, if the dusts are randomly oriented in space (i.e., no charge, no magnetic field, no sedimentation process in a gaseous environment) and if they are statistically spherical (circular projected surface), LMT can be used to *estimate* some of their scattering properties (like diffraction patterns). As an illustration of LMT application to plasma, the reader is invited to see Fig. 17 in Ref. [13]: a comparison between experimental data and LMT predictions for the modulus Δ and the polarization angle ψ of the photo-detector signal from an ellipsometrer measuring dusts produced in Ar–CH_4 plasma. The numerical simulations are found to be in good agreement with what was measured for dust precursors. The use of a LMT with a layered particle model or the T-Matrix with a cluster particle model could further improve the agreement with these experimental data [13,14].

The T-Matrix theory was first introduced by Waterman [15] has a method to calculate the scattering of an electromagnetic wave by a homogeneous and non-spherical particle [16]. The basic concept of this theory is to replace the scattering object by a set of surface current densities, so that in the exterior domain the sources and fields are exactly the same of those existing in the original scattering problem. Once the surface current densities are determined, the scattered field outside the circumscribing sphere is obtained by using the representation theorem. Finally, the transition matrix is obtained by assuming spherical vector wave expansions for the incident and the scattered fields. This allows determining the Transformation Matrix or 'T-Matrix' that links the scattered field to the incident field. All the scattering properties of a particle are described by its T-Matrix.

To calculate the T-Matrix of spheroids, truncated cylinders and 'Chebyshev' particles, we use the extended precision Fortran code developed in NASA Goddard [16]. Various scattering quantities properties can be deduced from this matrix: the cross-sections C_{ext}, C_{sca}, C_{abs}, the factor of symmetry, the polarization state... of the scattered light. Unfortunately, for numerical reasons, the maximum size-parameter ($\alpha = \pi D/\lambda$) cannot be large (depending on the wavelength λ, the refractive index, the particle shape and aspect ratios ξ), see Fig. 1. The computational time is also quite prohibitive so that the T-Matrix cannot be used for real-time inversion methods (it takes us one week to calculate the kernel matrix necessary to inverse data from Fig. 4). Thus our approach is to produce look-up tables of the T-Matrix results over a large range of parameters (refractive index, shape, aspect-ratio and wavelength). With

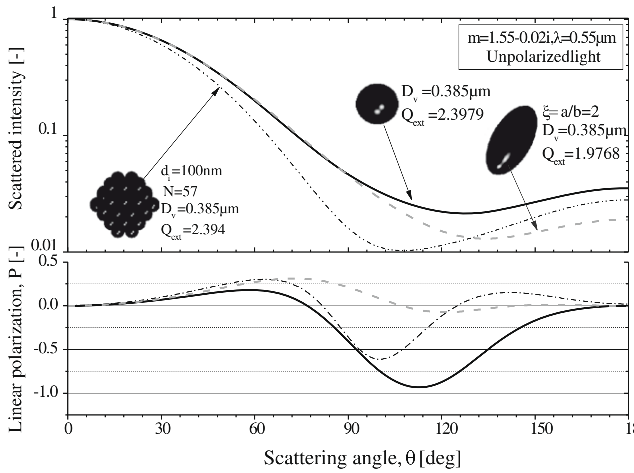

Fig. 2. Comparison of the scattering diagrams and the degree of linear polarization of a carbonaceous: sphere, ellipsoid and compact aggregate, with all the same equivalent volume.

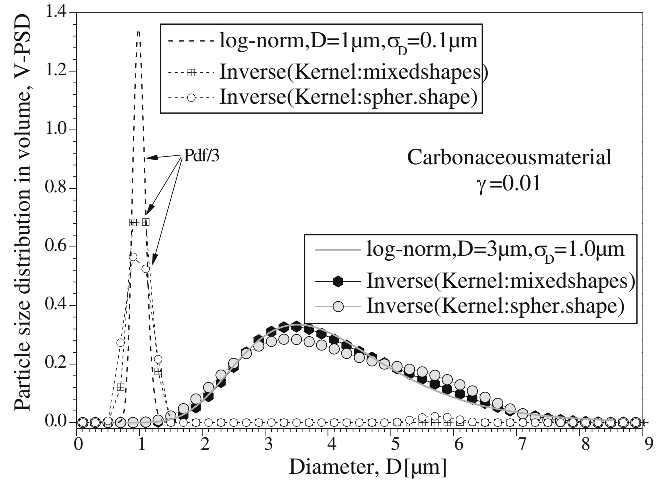

Fig. 4. Simulated and inverse size distribution: effect of dust shape on the quality of the reconstruction.

this database we can predict, in few seconds, all scattering properties of various clouds of particles with different compositions, shapes, aspect-ratio, etc. We mostly use the randomly oriented spheroid particle model as it allows breaking most of the contributions of SV-MDR [2]. The T-Matrix method can also be used to calculate the scattering properties of clusters, aggregates of spherical particles, compact or fractal [17]. For this purpose we have developed a software [14] that automatically generates and rotates clusters of particles, produces the corresponding batch files to run the T-Matrix code, produces statistical results and generates graphs automatically, etc... For the scattering of a 'cloud of dusts' the scattering properties are simply averaged over a large number of particles (i.e., multiple scattering effects between different clusters are negligible but they can exist inside the cluster itself). As an example of the particle shape influence on its scattering properties, Fig. 2 compares the scattering diagrams and the degree of linear polarization of a carbonaceous: sphere, ellipsoid and compact aggregate, with the same equivalent volume [2].

The Discrete Dipole Approximation (DDA) is a numerical method which solves the problem of the scattering and absorption by an array of polarizable point dipoles interacting with a monochromatic plane wave [5]. To do so, the particle model, whose shape can be totally arbitrary, is meshed with thousands or millions of finite elements. The DDA method requires intensive calculations and it is subject to increasing numerical errors for particles that are large compare to the incident wavelength or with a high refractive index. Accurate calculations of particles scattering properties can be obtained for $(2\pi|\tilde{m}|D/\lambda) < 0.5 \sim 1$. It means that with the code DDSCATT [18] for instance, and for accurate extinction cross-sections calculations, the diameter of carbonaceous particles is limited to $D = D \leqslant 0.9$ μm for $\lambda = 0.5$ μm and $D \leqslant 18$ μm for $\lambda = 10$ μm. As the accuracy of this method decreases also rapidly for $|\tilde{m} - 1| > 2$, [17], it cannot be used with confidence for the largest metallic dusts (We, Be) when they are observed in the NIR–MIR region [2].

Geometrical optics is known to be not appropriate to model pure wave phenomena and the scattering of particles with a diameter close to the wavelength [5], see Fig. 1.

3. Spectral extinction technique

3.1. Principle

Regarding to the constraints imposed for the situ characterization of dust (see Section 1), we have chosen to develop an optical diagnosis based on the measurement of the 'turbidity' of the vacuum vessel atmosphere [2]. The corresponding technique will be called 'light extinction spectrometry' further on as it shows strong similarities with the well known 'light absorption spectrometry'. Up to known this technique has been mostly used to characterize, in laboratory, liquid suspensions or small droplets in air [12]. However, some studies have been devoted to the sizing of crystals [19] or soot aggregates analyses [6,12]. The challenge here is to extend this technique so as it can satisfy the ITER needs: determination of dust's number concentration N and size distribution $f(D)$ over a long distance (several meters), only one or two optical accesses, particles having different and complex shapes and compositions [2].

Basically this technique requires to pass, through the cloud of particles to be analyzed, a collimated and polychromatic beam with spectral intensity $I_0(\lambda_i)$ and wavelengths λ_i, see Fig. 3. The transmitted spectral intensity $I(\lambda_i)$ is collected and directed towards a spectrometer. If the collection of multiple scattered photons is negligible [11], the beam transmission $T(\lambda_i)$ is given by:

$$T(\lambda_i) = \bar{I}(\lambda_i)/I_0(\lambda_i) = \exp(-\tau L), \quad (1)$$

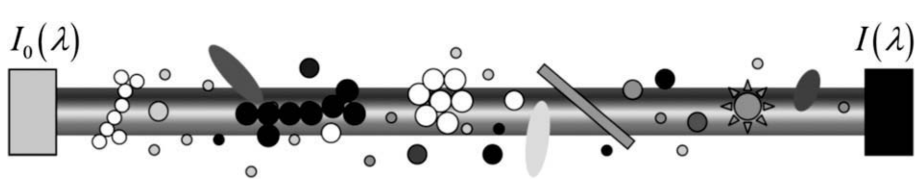

Fig. 3. Schematic of the principle of the extinction spectrometry.

where L is the path length of the beam through the cloud of dusts, $\tau = N\bar{C}_{\text{ext}}$ is the 'turbidity of the medium'. $\bar{C}_{\text{ext}}$ is an integral quantity which represents the mean extinction cross-section of the particles. With Q_{ext} being the extinction coefficient of a particle characterized by $(D, \tilde{m})$ we have

$$\bar{C}_{\text{ext}} = \int_{D_{\min}}^{D_{\max}} Q_{ext}(D, \tilde{m}) \frac{\pi D^2}{4} f(D) dD. \tag{2}$$

3.2. Inversion procedure

Mathematically, the previous equation is known as a so-called 'inhomogeneous Fredholm equation of the first kind'. Our problem is to find the particle number concentration N and the particle size distribution in number (N-PSD): $f(D)$, from the measured transmission $T(\lambda_i)$ and by using the calculated kernel $Q_{ext}(D, \tilde{m})$. This problem is predetermined, ill-posed, and requires a specific inversion procedure.

For numerical reasons, it is preferable to express the particle concentration in volume C_v rather than in number. For equivalent spherical particles the latter parameter is simply related to the particle number concentration by $V(D) = C_v v(D) = N(\pi/6)D^3 f(D)$, where $V(D)$ is the particle size distribution in volume (V-PSD) and $v(D)$, the normalized particle size distribution in volume. By replacing number quantities by volume ones and by introducing the constant $\Lambda = -3L/2$ we can rewrite Eq. (2) in the following linear form:

$$\ln[T(\lambda_i)] = \Lambda \int_{D_{\min}}^{D_{\max}} Q_{ext}(\lambda_i, r, \tilde{m}) \frac{V(D)}{D}. \tag{3}$$

This integral equation can be discretized as follows:

$$\int_{D_{\min}}^{D_{\max}} \frac{Q_{ext}(\lambda_i, \tilde{m}, D)}{D} V(D) dD = \sum_{j=1}^{M} S_{i,j} V_j, \tag{4}$$

where the vector to be determined is the the discrete form of $V(D)$, V_j with $j = 1, 2, \ldots, M$. The element $S_{\text{i,j}}$ is equal to $S_{i,j} = Q_{ext}(\lambda_i, \tilde{m}, D_j)/D_j$ for wavelength $\lambda_i, i = 1, 2, \ldots, N \cdot \mathbf{S}$ is a $N \times M$ 'extinction matrix' which has to be calculated only once with

$$S_{i,j} = \int_{D_{j-1}}^{D_j} \frac{Q_{\text{ext}}(\lambda_i, \tilde{m}, D)}{D} dD. \tag{5}$$

A measured transmission spectrum can be represented as a vector $\bar{\mathbf{T}}$ whose element $\bar{T}_i$ represents the beam transmission at wavelength λ_i by a particle cloud with size distribution $\mathbf{V}$. To find $\mathbf{V}$, we have to solve a linear algebraic equation: $\bar{\mathbf{T}} \equiv \mathbf{S} \cdot \mathbf{V}$. The solution of this equation can be written as: $\mathbf{V} \equiv \left(\mathbf{S}^{\mathbf{T}}\mathbf{S}\right)^{-1} \mathbf{S}^{\mathbf{T}}\bar{\mathbf{T}}$. In fact this solution is ill conditioned and numerically unstable. To overcome this problem one way is to minimize iteratively the square of the difference $\mathbf{S} \cdot \mathbf{V} - \bar{\mathbf{T}}$ and taking into account the fact that the PSD is always a positive quantity $(V_j > 0, j = 1, 2, \ldots, M)$. It turns to minimize a Non Negative Least-Square problem (NNLSQ): $\underset{V>0}{Min} \left\| \mathbf{S} \cdot \mathbf{V} - \bar{\mathbf{T}} \right\|^2_{2-LSQ}$. This minimization process can be performed by using orthogonal numerical algorithms [20]. Note that the present authors have used with success this method to inverse the critical scattering patterns produced by clouds of bubbles. However, extinction spectrometry deals with particles sizes ranging between the Rayleigh and the Mie scattering regimes. So that, extinctions coefficients can change of several order of magnitude from the lower to the upper boundary of the PSD, leading to extremely large condition number of the matrix $\mathbf{S}$. To limit this problem, we introduce a smoothing matrix H and a Lagrangian parameter (i.e., smoothing factor) [20] so that the algebraic equation becomes

$$\left(\mathbf{S}^{\mathbf{T}}\mathbf{S} + \gamma \mathbf{H}\right)\mathbf{V} = \mathbf{S}^{\mathbf{T}}\bar{\mathbf{T}}. \tag{6}$$

The problem now leads to minimize the following quantity:

$$\underset{V>0}{Min} \left\| \left(\mathbf{S}^{\mathbf{T}}\mathbf{S} + \gamma \mathbf{H}\right)\mathbf{V} - \mathbf{S}^{\mathbf{T}}\bar{\mathbf{T}} \right\|^2_{2-LSQ}. \tag{7}$$

As an illustration of typical results obtained with this inversion procedure, Fig. 4 compares the simulated log-normal V-PSD and the reconstructed distributions, for different particle shapes and kernels. These results were obtained for a wavelength range and resolution of, respectively, 0.4–10 µm and ≈ 0.1µm (i.e., N = 97 wavelengths and M = 100 size classes).

4. Conclusion and perspectives

Different numerical and theoretical tools have been developed to predict the various scattering properties of dust encountered in tokamak, and to inverse the Fredholm integral obtained with collective optical sizing techniques (extinction, diffraction, polarization, etc.).

Perspectives for this work will be to further improve the modeling of dust refractive index and shape, as well as to develop an extinction spectrometer to be tested on controlled aerosols of W, Be, C mixtures.

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