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# SIZE EFFECTS ON THE SCATTERING MATRICES OF CLAY PARTICLES: AN EXPERIMENTAL STUDY

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ABSTRACT. We present experimental scattering matrix elements as functions of the scattering angle of two sets of three samples of clays (yellow, green, and white). The measurements were performed in Amsterdam at a wavelength of 633 nm, and at the IAA cosmic dust laboratory in Granada at 647 nm. We study the impact of different sizes on the measured scattering matrix elements.

## 1. Introduction

One of the biggest difficulties in modeling light scattering by irregular dust particles is widely known. The intermediate size parameters between the resonance domain (sizes comparable to the wavelength of the incident light) and the geometric-optics domain (sizes much larger than the wavelength of the incident light) appear to present an extremely difficult task (e.g. [1]). A major advantage of experimental measurements at visible wavelengths is that we can deal with real ensembles of dust particles covering relatively broad size ranges (including the resonance region). However, a systematic study of the impact of different physical properties on scattering of the sample under study remains extremely challenging due to the difficulty in finding the appropriate samples. Several attempts have been previously made to experimentally study the size effect on the scattering behavior of irregular dust particles [2]-[4]. In those studies the original bulk samples were *ad hoc* prepared with elaborate sieving procedures so that the measurements could be repeated for different size distributions. Despite all efforts, the final size distributions were remarkably similar for size parameters smaller than about 10. In this work we present scattering matrix measurements as functions of the scattering angle for two sets of three clay samples (yellow, green, and white). The measurements were first performed in the Amsterdam light scattering laboratory [5]. Those for green clay have been previously published [6] but that is not the case for white and yellow clay. About ten years later, we have found significant variations in the size distributions of the yellow and white clay samples. This put us in the unique position to study the effect of small differences in the size distribution on the scattering matrix of clay particles by performing new measurements at the IAA Cosmic Dust Laboratory in Granada [7].

## 2. Description of the samples

The particles of the three clay samples present a sheet-like structure. As an example in Fig. 1 (left panel) we present a Field Emission Scanning Electron Microscope (FESEM) image of yellow clay particles as they are during the light scattering measurements. A FESEM slide is used to collect particles directly in the jet stream of the aerosol generator by holding the slide briefly in the jet at the place where it intersects with the laser beam. The exact values of the refractive indices of the three clay samples are unknown. Based on literature values, we can assume that the real part of the refractive index lies between $1.5$ and $1.7$, while the imaginary part likely lies in the range between $10^{-2}$ and $10^{-5}$ at visual wavelengths.

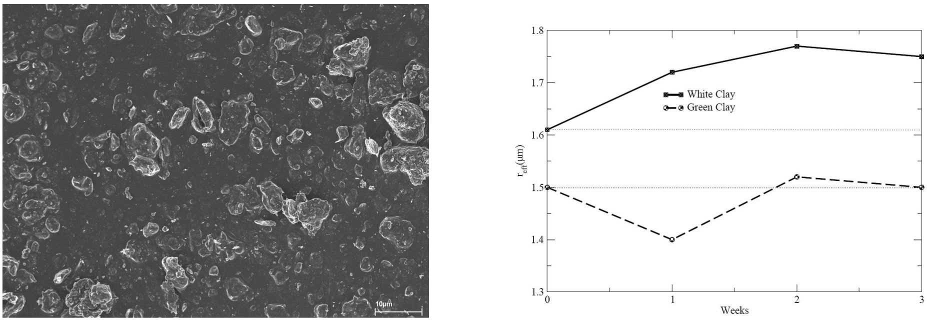


**Figure 1.** FESEM image of yellow clay directly collected from the aerosol jet stream. The white bar at the bottom right corner denotes $10\mu$m (left panel). Evolution of the effective radii of white (top) and green clay (bottom) over three weeks (right panel).

The projected surface area distributions of the three clay samples were measured in Amsterdam by using a Fritsch laser particle sizer [9] that employs the Fraunhofer diffraction theory for spheres. About ten years later, the size distribution measurements of the three samples have been repeated in Granada with a Mastersizer2000 from Malvern instruments. To retrieve the size distributions we selected the Fraunhofer diffraction mode. In Table 1 we present the values of the effective radius, $r_{\text{eff}}$, effective size parameter $x_{\text{eff}} = 2\pi r_{\text{eff}}/\lambda$, and effective variance, $v_{\text{eff}}$, obtained from the measured size distributions in Amsterdam and Granada, respectively. Each (Granada) size distribution data presented in this abstract is based on an average of 6 independent size distribution measurements. Corresponding standard deviations are also provided. We should keep in mind that small differences in the size distributions measured in Amsterdam and Granada could be due to the fact that they have been measured with completely different devices. Nevertheless, we have found differences as high as $11\%$ and 25 $\%$ in the retrieved $x_{\text{eff}}$ for yellow and white clay, respectively. It is well known that the water uptake and swelling ability is different depending on the type of clay (see e.g. [8]). To check whether ambient conditions like atmospheric humidity could have played a role we have performed the following test at the two extreme cases, namely green clay and white clay. A certain amount of white clay particles have

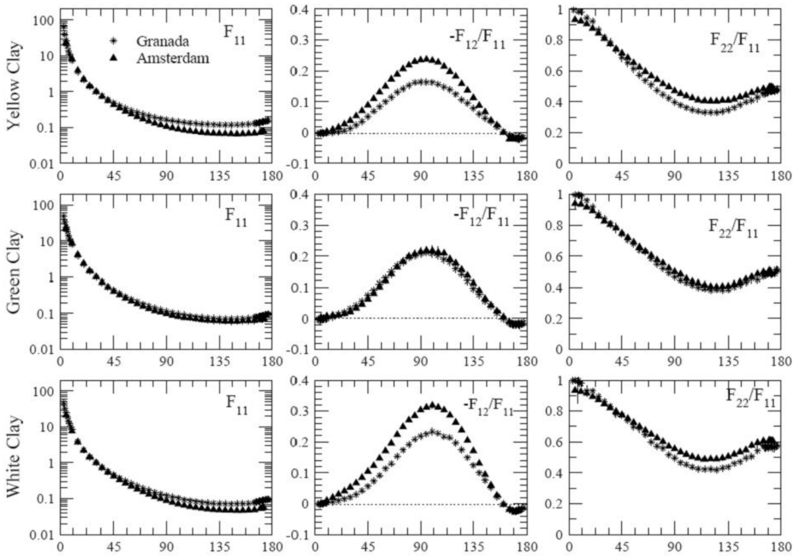


**Figure 2.** Measured scattering matrix elements as functions of the scattering angle, $\theta$, in Amsterdam (asterisks) and Granada (solid triangles). Top, medium, and bottom panels correspond to the measurements for the yellow, green, and white clay samples, respectively. Errors are presented by bars.

been put in a small pot. We have left the pot open for three weeks, performing size distribution measurements every week. The $r_{\mathrm{eff}}$ of white clay increased during the first two weeks from its original value, suffering a slight decrease in the third week. In total the $r_{\mathrm{eff}}$ of the white clay sample increased 0.14 $\mu$m in three weeks. During the same three weeks the same test was performed with green clay. In this case the $r_{\mathrm{eff}}$ fluctuated around its original value slightly decreasing in the first week increasing again in the second and third week up to the original value. The results of the test are presented in Fig. 1 (right panel). Thus, it seems like the size distribution of white clay may indeed have been affected by the ambient conditions whereas the green clay is hardly affected by it.

**Table 1**

| Sample | $r_{eff}$ A | $r_{eff}$ Gr | $x_{eff}$ A | $x_{eff}$ Gr | $v_{eff}$ A | $v_{eff}$ Gr |
|---|---|---|---|---|---|---|
| Yellow | 1.67 $\mu$m | 1.82 $\mu$m $\pm$ 0.00 | 16.6 | 17.7 | 3.4 | 2.93 $\pm$ 0.03 |
| Green | 1.55 $\mu$m | 1.62 $\mu$m $\pm$ 0.01 | 15.4 | 15.7 | 1.4 | 1.55 $\pm$ 0.02 |
| White | 1.26 $\mu$m | 1.57 $\mu$m $\pm$ 0.02 | 12.5 | 15.2 | 2.1 | 1.42 $\pm$ 0.02 |

## 3. Measurements

In Fig. 2 (left column) we present the phase functions, $F_{11}(\theta)$, measured in Amsterdam (at 633 nm) and Granada (at 647 nm) normalized to one at 30 degrees scattering angle. In the center and right columns we present the degree of linear polarization for unpolarized incident light ($-F_{12}(\theta)/F_{11}(\theta)$) and the $F_{22}(\theta)/F_{11}(\theta)$ ratio, respectively. Top, medium and bottom panels correspond to yellow, green, and white clays, respectively. We refrain from showing the other measured scattering matrix elements since they are not significantly affected by the differences in the size distributions. The measured scattering matrix elements will be available in digital form in the Amsterdam-Granada light scattering database at www.iaa.es/scattering. Although the scattering matrices of the green clay sample have been measured in Amsterdam and Granada so far away in time and with a different experimental apparatus in a different laboratory, the agreement in the measured results is impressive. The small differences found in their size distributions are not large enough to produce a significant effect in the measured scattering matrix elements. In contrast, there is a clear tendency in the measured $-F_{12}(\theta)/F_{11}(\theta)$ for yellow and white clay. The smaller the size parameter [Cf. Table 1] the larger the maximum of the degree of linear polarization. The results for the $F_{22}(\theta)/F_{11}(\theta)$ ratio exhibit larger values from around 45 degrees scattering angles for the samples with smaller size parameters. Thus, an increase in size seems to produce a decrease in the minimum of the $F_{22}(\theta)/F_{11}(\theta)$ ratio. We also see a size effect in the measured phase functions as functions of the scattering angle. The samples with larger size parameters show larger values at side- and back-scattering angles. In summary, we found that the size distributions can change in time. But the results for the green clay show that, if the size distribution remains nearly the same, so do $F_{11}(\theta)$, $-F_{12}(\theta)/F_{11}(\theta)$ and $F_{22}(\theta)/F_{11}(\theta)$. Therefore, conceivable other changes, e.g. in composition, shape and structure, do not occur or are not important for the light scattering matrix. This holds at least for the green clay samples and, presumably, also for the other clay samples considered in this paper.

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