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New perspectives for the understanding of the optical properties of middle-age nano-cermets: The lustres

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Abstract

Lustres are famous for their shinning metallic colours displayed in the specular direction. The optical phenomena that occur in this direction are plasmon absorption and interferences, linked with the multi-layer structure of the lustre. In order to understand the optical properties of lustre, we need to study also the other directions. Doing this, we discovered that coherent phenomena still play a key role around the specular direction leading to a smooth transition from the colour observed in the specular direction to the one observed for the diffused reflection. These coherent phenomena are identified as interferences that do appear away from the specular direction and assumed to be linked with the roughness of the interfaces of the multi-layer structure.

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1. Introduction

Lustre: This word refers to a large range of potteries created between the 9th and 17th century in various locations of the Mediterranean basin, from Middle East to Spain and Italy. The particularity of these potteries is their surface decoration [1]. Craftsmen of Middle Age and Renaissance times managed to produce at the surface of the glaze covering the pottery, using complicated firing processes, nano-particles of noble metal (silver and/or copper) [2]. The presence of these nano-particles at the surface of a pottery is sometimes considered enough in order to classify it as lustre [3,4]. But from an historical point of view it cannot be the case. In fact, the most astonishing effect of lustres is the changing of colours observed in the specular direction, the pottery displaying shining metallic iridescent colours, due to the presence and the organization of these metallic particles at the surface.

This effect was the one search by the craftsmen and it is for this effect that lustre became one of the most famous decorative objects at that time. Of course, the craftsmen of these centuries did not know they were producing nano-particles. That is why it is preferable to consider the optical properties of these objects in order to give a definition of lustre. The potteries not displaying the shining effect but comprising nano-particles are then considered as failures.

The optical properties observed in the specular direction are due to non-independent phenomena (plasmon absorption, interferences, diffusion) as shown in previous studies [5,6]. However, until now, no studies have been carried out in order to understand how the distribution of the nano-particles will influence the optical properties of the lustre in any other direction. While studying this problem, other questions arise. How the transition occurs, from the optical point of view, between the colour observed in the specular direction and the colour in any other direction? Is it a sharp transition? Are there new phenomena to take into account? In order to answer these questions, we performed a series of measurements of reflection spectra in various directions. The results of these measurements and their discussion are the purpose of this paper.

In the next part the measurements are presented, followed, in a third part, by their comparison with some

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simulations. Then, a new hypothesis is set out in order to understand the optical properties observed and this hypothesis is discussed in the last part. As a conclusion, we derive, from this work, new optical properties that could be included in the definition of lustre.

2. Gonio-spectro-photometric measurements

The instrument used for the measurements is a gonio-spectro-photometer whose description is given in Fig. 1.

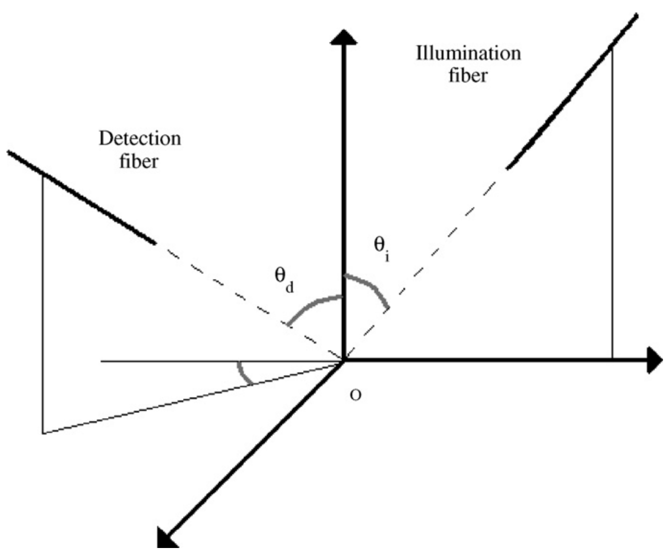

Fig. 1. Schematic view of the gonio-spectro-photometer.

It is composed of two fibres. The illumination fibre can be orientated so that it is possible to manually change the angle of incidence θ_i. The detection fibre is placed on two motorized arms that can rotate in two directions, leading to the angle θ_d and φ, so that it is possible to perform measurements in any direction. With this device, it is possible to measure spectra of reflection away from the specular direction ($\theta_d \neq \theta_i$ and/or $\varphi \neq 0$).

Such measurements were performed on a lustre named L2rob, from Italy, Renaissance time, whose structure is one layer containing silver nano-particles, covered by a glassy layer. The spectra measured are displayed in Fig. 2. The measurements are performed in the plane of incidence ($\varphi = 0$) for various couples of angles θ_i and θ_d, the label of the spectrum being $|\theta_d - \theta_i|$. On this figure, we see that as the spectrum is measured further from the specular direction, the intensity of the first peak, at around 375 nm, decreases whereas no such behaviour is observed at larger wavelengths for the second maximum.

3. Simulations

In order to simulate the spectra measured in non-specular directions, we used a diffusion model: a 4-flux model. In this model, the transports of energy in the medium are represented by four fluxes, a collimated and a diffuse flux propagating in the incident direction and other collimated and diffused flux propagating in the other direction. Collimated fluxes are absorbed by the medium and transformed into diffused flux by diffusion phenomena.

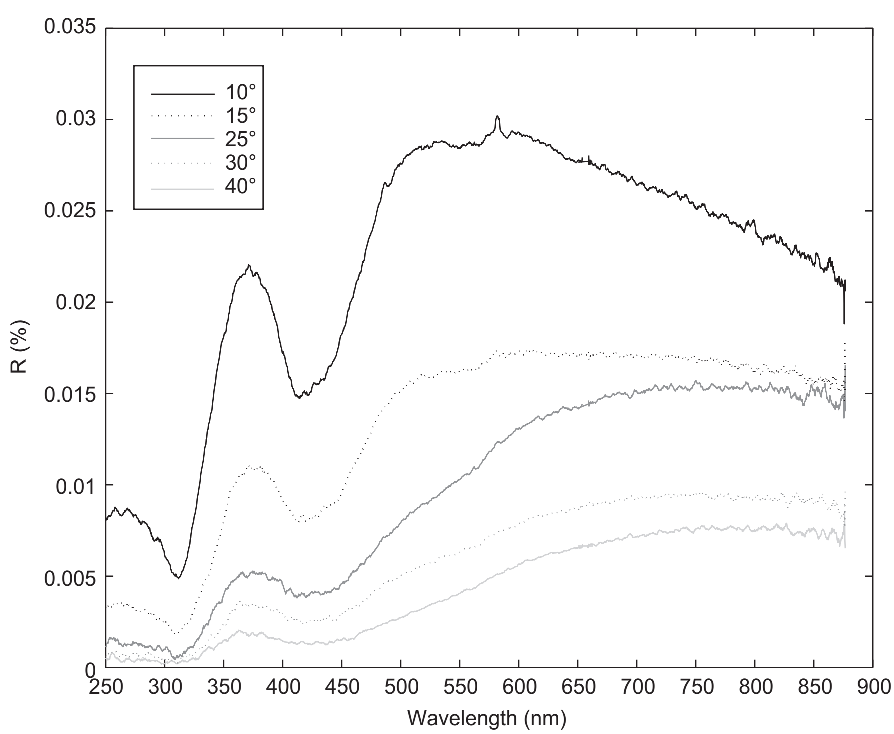

Fig. 2. Reflection spectra measured in non-specular direction (diffused reflection) on shard L2rob, the angle mentioned is the angular distance from the specular direction.