

INFLUENCE OF THE SUBSTRATE COLOUR ON THE VISUAL APPEARANCE OF GILDED SCULPTURES

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KEY WORDS: colorimetry, spectrophotometry, SEM, image synthesis, materials sciences, archæology, restoration, metallurgy

ABSTRACT:

Measurements, optical simulations, samples realization of gilded surfaces were made for studying the visual appearance and the substrate color influence of gilded surfaces mainly encountered in sculpture. The presented study concerns the characterization of the extrinsic and intrinsic parameters useful for the optical simulation of the visual properties of the gilded surfaces. Thus, geometrical and optical data have been acquired. These measurements on several cross-sections give the characteristic thickness of a sample, very useful for the spectral simulations. The mean geometrical parameters are completed by laser profilometry measurements made on the prepared samples. We have obtained the optical characteristics of pure gold (bulk sample) and some alloys of gold by spectroscopic ellipsometry and obtained very detailed complex indices of refraction. It appeared that the gold-leaf were about $2\mu\text{m}$ thick. Such a thickness allows a fraction of the incident light to be transmitted through the gold-leaf and to reach the gold-size. That weak spectral transmissivity is then coupled with the scattering of the incident light due to the small cracks or holes appearing in the so mechanically fragile gold-leaf. Thus, fundamental mechanisms of light-matter interaction are simultaneously present as optical transmissivity by the metallic film, surface plasmons and very small holes, macroscopic holes and cracks. The spectrophotometric measurements also give access to a statistical estimation of the holes and diameters densities extracted from SEM or optical microscopy imaging. These quantitative informations are obtained by comparison between bulk and gold-leaf reflectance factors at normal and oblique incidence. The computed images based on the measured optical properties on all the materials of the sculptures are obtained thanks to an adapted model from Cook-Torrance's one. As a side effect, a surface plasmon resonance contribution is added for understanding the spectral transmissivity through the gold leaf. The specular reflection on the rough metallic interface is also enriched by the volume scattering originating in the pigmented layer made by the gold-size. That scattering contribution is modeled by the phenomenological Kubelka-Munk formulas. Then rendering is finally made with our spectral ray-tracer called Virtuelium(CALLET and ZYMLA, 2006).

1 INTRODUCTION

We have led many experiments for characterizing the real influence of the substrate coloured paint, called gold-size, on the global visual appearance of several gilded surfaces. Many works of art are covered by a gold film deposited by different processes. Among these are mercury gilding(BOWMAN et al., 1997) and gold-leaf gilding. The substrate colour is generally depending on the material in which the work of art is made. The reasons are found in the mechanical properties needed for fixing the noble metal on the material support. We have studied gilded sculptures or ornaments used in architecture and we will present the complete scientific chain of treatment for a virtual or real restoration. Gilded brasses obtained by mercury gilding and their corrosion influence will be shown thanks to scanning electron microscope (SEM). For metallic sculptures when made of gilded brasses for example, a very thin crack in the gold-leaf causes the corrosion process to start and to recover an important part of the object. The optical appearance of such a gilded surface is then deeply modified. The laser cleaning, if suitably adjusted in power, can reveal the original aspect.

The presented study concerns the characterization of the extrinsic and intrinsic parameters useful for the optical simulation of the visual properties of the gilded surfaces. Thus, geometrical and optical data have been acquired. These measurements on several

cross-sections give the characteristic thickness of a sample, very useful for the spectral simulations. The mean geometrical parameters are completed by laser profilometry measurements made on the prepared samples. We have obtained the optical characteristics of pure gold (bulk sample) and some alloys of gold by spectroscopic ellipsometry and obtained very detailed complex indices of refraction(CALLET et al., 2002). It appeared that the gold-leaf were about $2\mu\text{m}$ thick. Such a thickness allows a fraction of the incident light to be transmitted through the gold-leaf and to reach the gold-size. That weak transmittivity is then coupled with the scattering of the incident light due to the small cracks or holes appearing in the so mechanically fragile gold-leaf. Considering wood or stone sculptures, the substrate is not a metallic and specularly reflecting surface but generally offers a reddish-brown colour, modified by the optical properties of the gold-size, an oil size of damar and red or yellow ochres. The optical influence of the first layer deposited on the sculpture itself is negligible when the coloured layer is present. A black paint is also encountered on white marble sculptures (e.g. asian antique art) just behind the gold-leaf. A visual comparison between samples obtained with different methods of preparation is possible and we shall show how a spectral model of rendering in realistic image synthesis can help to give a plausible vision of a previous and original state of a gilded sculpture. A Middle Age statue with some gilded parts has been studied (the recumbent statue of Philippe-Dagobert, 1222-1234) for a complete work on

the medieval polychromy restitution. For that project some samples have been prepared according to the medieval materials and techniques used for the gilding of stone. The samples were prepared on the same stone than the original statue, a fine limestone extracted from the open quarry of Saint-Maximin (40km in the north of Paris). The sample preparation consists in the deposition of the first layer of lead white and above, after drying, the gold-size itself and last the gold-leaf. For each step a spectrophotometric measurement is made all over the visible spectrum and more for the range [200 ; 900] nm. The influence of the lighting conditions is also presented. This is important today because the lighting systems used in the museum are very different of the lighting conditions where the work of art was originally exposed. If we want to place a gilded work of art in its historical environment, when it is reasonably possible, we have to retrieve a typical and plausible lighting. As an important result we shall exhibit the role of the polishing of the gold-leaf. The small cracks and holes made by this polishing process leave a non-negligible fraction of the incident light to be backscattered and then superimposed to the regularly reflected light. However slightly modified by the own roughness of the gold-leaf. Hence a surface scattering due to the roughness of the metallic surface is added to an internal scattering taking place inside the gold-size layer itself. Though this effect is not quantitatively important it offers a visual impact not negligible at all. The spectrophotometric measurements also give access to a statistical estimation of the holes and diameters densities extracted from optical microscopic imaging. These quantitative informations are obtained by comparison between bulk and gold-leaf reflectance factors at normal incidence.

2 SAMPLES PREPARATION

There are two main processes for gilding of wood and stone. We shortly describe here these processes. Water gilding is the traditional gilding of wood. A thin layer of white or red gesso mixed with rabbit skin glue is applied to the wood. A water-based bole is applied, the color of which will affect the color of the finished gilding. The bole is wet with gilder's liquor, a combination of alcohol and water which brings the glue up to the surface of the bole. Metal leaf (most commonly gold or silver leaf) is applied using a gilder's tip, a brush which is usually made of ox hairs. The leaf is later burnished with a tool using a semi-precious agate stone. Oil gilding is the traditional process for building exteriors. After the surface is cleaned and primed an oil "size" - similar to varnish - is applied. The size cures until it comes to tack (that is, it has dried enough to hold the leaf on to the surface but is still "tacky" - sticky - enough for the leaf to adhere to it). The surface is then skewed to fully press the leaf against the size and also remove any loose fragments of leaf. Oil gilded surfaces are not burnished any further.

We have prepared several samples of gilded surfaces following the traditional technique used for gilding on stone. As one of studied object was the recumbent statue of Philippe-Dagobert (1222-1232), a fine limestone sculpture, partly gilded, we made experiments on the state of surface of the gold leaf deposited on different layers colored or not. Thus, a first layer of white lead filler ($PbCO_3 \cdot Pb(OH)_2$), a second layer of colored preparation with a mixing of cinnabar (HgS) with red lead (Pb_3O_4) and called the gold-size or "bole", the third layer being made of an oil size only useful for fixing the gold leaf, were deposited. Some samples, when completely dry, were polished according to the medieval technique. This process reduces the surface irregularities increasing the smoothness and consequently the surface shininess while it introduces some holes by local abrasion and cracks of the gold leaf. The gold leaves themselves were 8 cm x 8 cm (24 karat, pure gold) and about 200 nm thick. As we

mentioned the colour of gold alloys is a subtle visual effect produced by polished surfaces. The table 1 below gives the classical denominations of such chromatic effects.

Gold Denomination	Gold	Copper	Silver	Iron
blue	75	-	-	25
grey	94	-	-	6
white	75	10	15	-
pink	75	5	20	-
red	75	25	-	-
green	75	-	25	-

Table 1: The conventional denomination of gold alloys.

3 OPTICAL MEASUREMENTS

Among all the measurements made in this study an important place was given to the extractible informations from the visible domain. The composition of gold alloys greatly influences the visual appearance and colour. In the medieval era the elementary composition of the gold leaves was approximatively of 56 to 97 % of gold and the complementary metal was copper. Thus, the bulk material could appear reddish enough to mask or reinforce the expected influence of the chromatic bole. In this study we restricted our work to pure gold leaves and eliminated the intrinsic alloys optical properties.

3.1 Optical imaging

We made many optical images at different magnifications for using in conjunction with laser profilometry measurements and for rendering in spectral ray-tracing.

3.2 Laser Profilometry

Laser profilometry is useful for the determination of the typical parameters of the holes and cracks made by the application process of finishing. The observed cracks and holes on our gilded samples have a mean horizontal extension of 300 nm and 60 nm depth (see figs. 1, 2) with covered areas of about 18 000 μm^2 . An attentive observation can help in the definition of the crater-like form of the holes. These geometrical indications are not sufficient for characterizing the visual appearance of the viewed surface. At a smaller scale a roughness is introduced even if the surface is burnished with an agate stone tool.

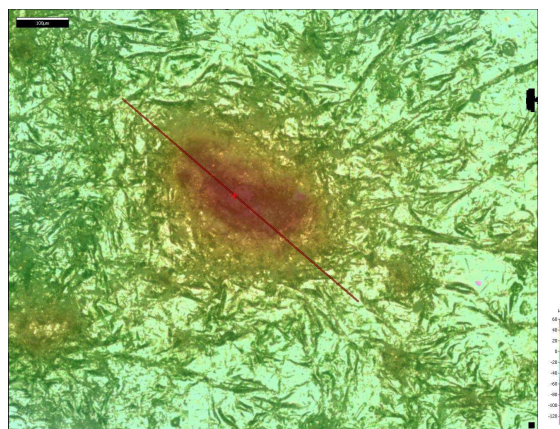


Figure 1: A sample of gilded surface where the gold size appears distinctly. Notice the line indicating the direction where the profile was extracted and drawn in fig.[2].

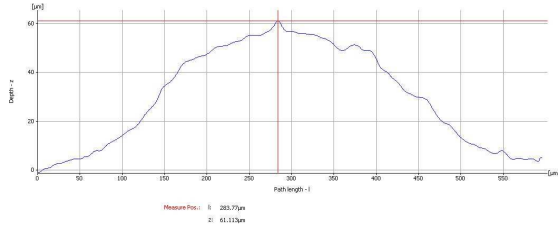


Figure 2: Typical profile measured by laser profilometry and corresponding to the line and cross drawn in fig.[1].

3.3 SEM imaging

The Scanning Electron Microscope gives very detailed informations on the state of surface at very small scales (see figures 3,4,5) but could not completely help to distinguish real holes that are missing zones of gold from craterlike cavities. These images are used to evaluate a statistical map of elevation and holes distribution (height fields) relatively to a reference altitude given by a mean level extracted from the images themselves. It is gener-

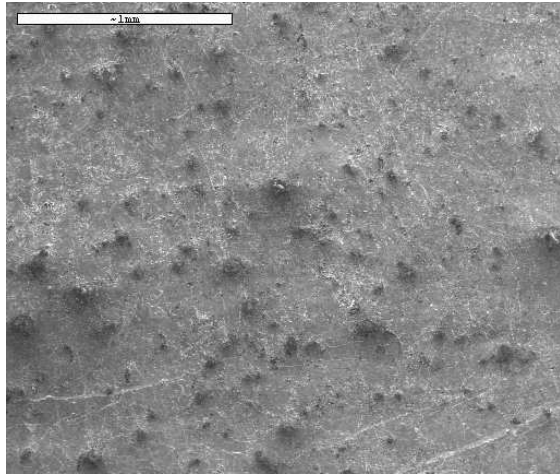


Figure 3: SEM image, at scale 1mm, where the global distribution of holes appears. Each hole, a black region craterlike, is about 0.1 to 0.3 μm large. The approximative covered area is 1.5 to 3% of the whole in a characteristic zone of the studied surface.

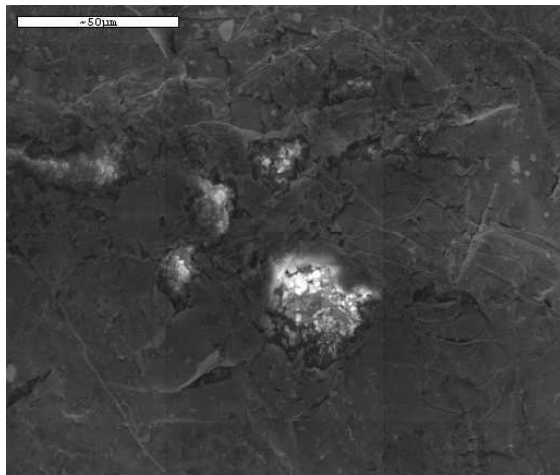


Figure 4: SEM image, at scale 50 μm , exhibiting the typical shape of the holes and the diffuse reflection areas from the bole.

ally admitted that the oil gilded surfaces are never mechanically burnished. We however polished some parts of the samples for having more complete optical measurements.

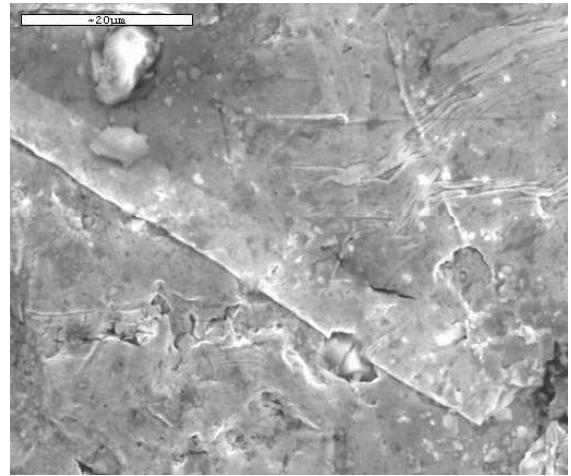


Figure 5: SEM image at scale 20 μm exhibiting the reinforcement of the reflectance factor due to the superimposed metallic gold film produced by the application process and the final burnishing.

3.4 Spectrophotometry

For understanding the visual influence of the gold-size and for estimating quantitatively the coloration of the reflected light emergent from a gilded surface, we made many measurements thanks to an Ocean Optics USB 2000 spectrophotometer. Among these measurements we shall only describe here the observations concerning the reddish gold-size with and without the gold leaf. The modelling of backscattering of light is mainly pertinent for diffusing materials. For very specularly reflecting materials, as polished metals are, the only backscattering of an incident light is due to roughness. If we suppose that the small holes and cracks, almost homogeneously distributed over the surface, bring a non-negligible contribution to the visual appearance, the backscattered spectrum must include an enhanced contribution in the red region of the visible spectrum. As a first visual experiment we have measured the backscattered light at normal incidence on non-burnished samples with and without coloured gold-size (bole). A subtle difference is visible on the spectra but are not very visually detectable for ordinary viewing conditions. The results of such a preliminary observation/measurements is given in fig. [6]. It appears that a small fraction of light is transmitted through the gold leaf and selectively absorbed by the reddish-brown gold-size. The mechanism by which such an absorption occurs could be a plasmon effect due to the dielectric-metal interface.

4 SIMULATION

The formulated model is derived from the classical Cook-Torrance model with roughness parameters. The proposed algorithm is briefly described hereafter. For the electronic plasma contribution (surface plasmons) modifying the spectral transmissivity of the gold leaf, we have calculated an effective index of refraction of the gold leaf (ABELES, 1963) according to a model of effective medium, shortly described now and derived from Maxwell Garnett theory (GARNETT, 1904). Let us consider a set of metallic inclusions of volumetric concentration p embedded in a homogeneous dielectric medium as described in fig. [8]. We successively

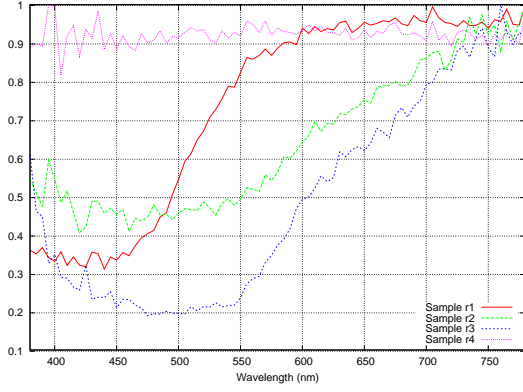


Figure 6: Comparison of the backscattering normalized reflectance factors spectra for the successive layers at normal incidence on four samples (r1) gold+oil size+bole+white lead, (r2) oil size+bole+white lead, (r3) bole+white lead, (r4) white lead.

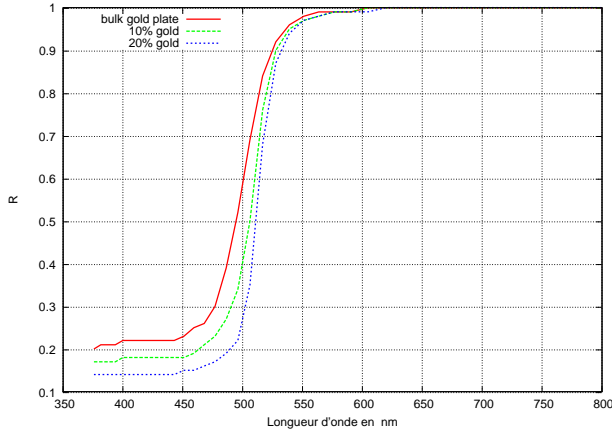


Figure 7: Comparison of the backscattering normalized reflectance factors spectra for the successive layers at oblique incidence on four samples (r1) gold+oil size+bole+white lead, (r2) oil size+bole+white lead, (r3) bole+white lead, (r4) white lead.

calculate the effective electric field acting on such a metallic inclusion. In reaction to this excitation the particle modifies the applied electric field and the electric displacement too. This mechanism leads to an effective dielectric function ε_e depending on the geometrical form of the metallic inclusions. The optical constants of gold are available in articles, books, metal handbooks ((SCHULZ, 1954), (CALLET, mars 1998)). Let us also consider, for generality, some lenticular particles having their short ellipsoidal axis c perpendicular to the horizontal delimiting faces of the global medium(BOHREN and HUFFMAN, 1983) ; the two other axes are such $a = b$ parallel to the horizontal interfaces. Viewed from above such a system could be described as a "go ban" with a unique kind of stones and where the go ban itself is the hosting dielectric medium. We define the excentricity as

$$e^2 = \frac{a^2}{c^2} - 1 \quad (1)$$

and the depolarization factor

$$A = \frac{1 + e^2}{e^3} (e - \tan^{-1} e) \quad (2)$$

Being given the two complex dielectric functions of gold and that (approximated) of the hosting medium, i.e. the oil size, we have,

the following *complex effective dielectric function* :

$$\varepsilon_e = \varepsilon_m \frac{\varepsilon_i [A + p(1 - A)] + \varepsilon_m (1 - p)(1 - A)}{\varepsilon_i A(1 - p) + \varepsilon_m [1 - A(1 - p)]} \quad (3)$$

All the ε depending on wavelength, we used the definition of the *complex effective index of refraction* $\hat{n}_e(\lambda)$ such that :

$$\hat{n}_e(\lambda) = \sqrt{\varepsilon_e(\lambda)} \quad (4)$$

In this last equation, the magnetic permeability was considered equal to unity as it is easy to verify in the visible domain. The reflectivity and transmissivity functions governed by the Fresnel formulas mainly depends on the imaginary part of the index of refraction defined in eq.4. That effective index of refraction(COHEN et al., 1973) has a lower imaginary part than pure gold and, consequently its spectral transmissivity is magnified. By this way the fraction of light diffusely emerging through the metallic leaf and originating in multiple scattering inside the volume of the bole layer is accessible. The effective medium formed by the metallic film, the nanoscopic holes and the oil size is approximated by a Maxwell Garnett model of the dielectric function[(ELIAS and LAFAIT, 2006)]. In eq.4 the optical absorption depends on the concentration p . R , the reflectance factor and p vary in opposite way. The upper value of p is fixed while no percolation process appears inside the effective medium made of holes and gold plates. ε_m is simply computed with an effective

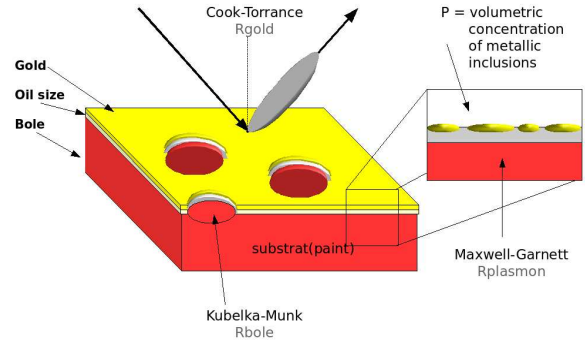


Figure 8: Schematic description of the proposed model with three chromatic components.

index of refraction for linseed oil used as oil size, i.e. $n_{oil} = 1.3$ Using the diffuse reflectance factor spectrum of the bole, considered as a completely opaque and multiple scattering pigmented medium, we add this contribution to the global reflectance factor. The main fraction of the reflected light is governed by Fresnel laws modulated in amplitude by an optical roughness. This term is the only classical Cook-Torrance one. The roughness influence is extracted from the profilometric measurements. The diffuse reflectance factor emerging through the metallic leaf and from the bole is obtained by the Kubelka-Munk model for opaque layers whose parameters (the K/S factors) are inverted from the diffuse and measured reflectance factor spectrum R_∞ of the only bole (sample "r3" in fig. 6 and 7).

$$\frac{K}{S} = \frac{(1 - R_\infty)^2}{2R_\infty} \quad (5)$$

The ratio defined in eq. [4] is important for adjusting the mixing of pigments while it is possible to use R_∞ directly for tests.

4.1 Proposed Algorithm

The previously mechanisms of light-matter interaction are then abridged in 3 components. In this set of interactions, the two macroscopic components are the most classical. The summation of the three independent mechanisms gives the received luminance. Thus, the regular reflection on the moderately rough gold surface, R_{gold} and the diffuse reflection from the missing parts from the bole R_{bole} is added to the plasmon surface effect $R_{plasmon}$. The Cook-Torrance model modulated by the optical roughness of the metallic film is used to compute R_{gold} thanks to the complex index of refraction given in (CALLET, mars 1998). The Kubelka-Munk model is used for R_{bole} . The last term $R_{plasmon}$ is more complicated. The effective interaction area is used for weighting these three terms.

$$R = fR_{bole} + (1 - f)((R_{gold} - T_{pgold}) + R_{plasmon}) \quad (6)$$

where f describes the ratio between the cumulated hole area and the global gold leaf area. T_{pgold} defines the gold transmissivity due to surface plasmon resonance. We thus consider an effective dielectric-metallic medium made of a metallic continuum embedding the holes in which an also effective resin is present and composed by the oil size and air. The model described here leads to the computation of an effective index of refraction for the surface plasmon component depending on the holes concentration. Two estimations for 10 % and 20% are presented in fig. [9,10]. One can observe on these figures that a slight red shift, depending on the holes concentration, in the visible spectrum is distinguishable. The modify reflectance factor of such a system is shown in fig. 11

The computed reflectance factor is used in our simulations for

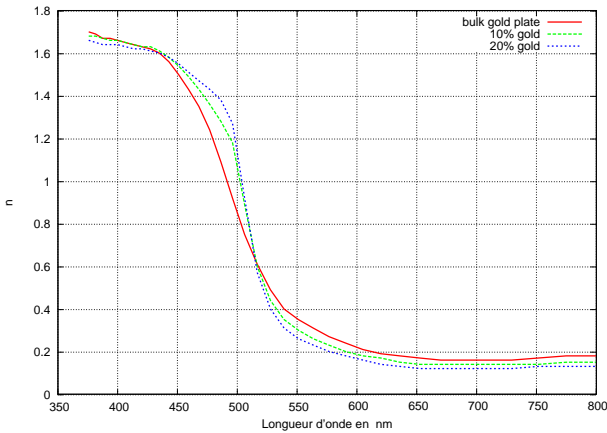


Figure 9: Comparison between the real parts of the index of refraction of a bulk gold plate and two effective gold films with 10% and 20% holes.

computing the received luminance by the virtual CIE standard observer.

5 RENDERING

The computed images based on the optical properties measured on all the materials of the sculptures are obtained with a model adapted from Cook-Torrance's model for the specularly reflection on the rough metallic interface enriched by the volume scattering originating in the pigmented layer made by the gold-size. That scattering contribution is modeled by the phenomenological Kubelka-Munk formulas. About the gilding on stone, as the colour studies led on the recumbent statue of Philippe-Dagobert,

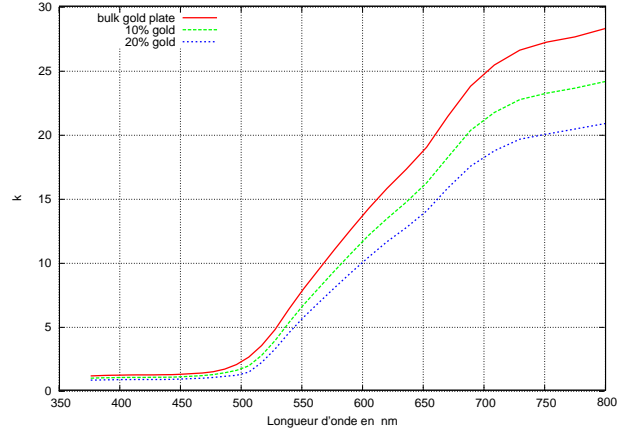


Figure 10: Comparison between the imaginary parts of the index of refraction of a bulk gold plate and two effective gold films with 10% and 20% holes.

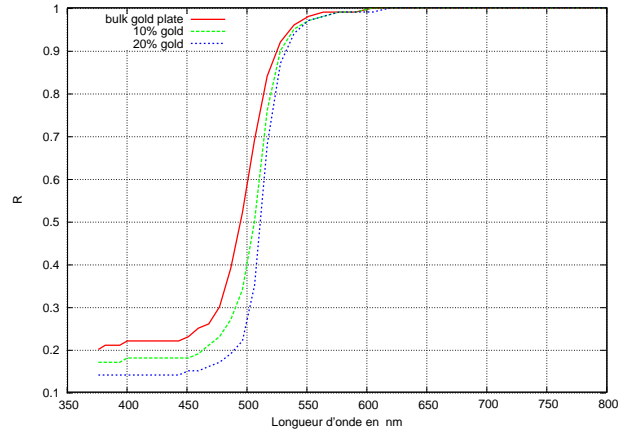


Figure 11: Comparison between the reflectance factors of a bulk gold plate and two effective gold films with 10% and 20% holes.

we publish our model on the small lion with pure opaque gold (fig. 12), gold leaf on a black bole (fig. 13) and gold leaf on cinnabar bole (fig. 14). The lighting conditions for all the computations are CIE D65 illuminants and a colorimetric observer CIE 1964 (10°).

6 RESULTS AND DISCUSSION

All the descriptions available in the scientific and artistic literature mention a visual influence of the coloured under-layer. We have experimented in two worlds if such an effect was measurable making assumptions on the materials structure and nature. It seems that fundamentals phenomena are responsible for the visual aspect of gilded surfaces. Plasmon surface and enhanced optical transmissivity due to an effective complex index of refraction of the gold leaf is then the most fundamental effect encountered. The validation of such a model is not very easy to obtain. The dual notion of "complementary material", defined on the basis of the effective media theories, has to be further explored.

7 CONCLUSIONS AND FUTURE WORK

We have to continue the work presented here by leading optical measurements on several stone sculptures and finding a better

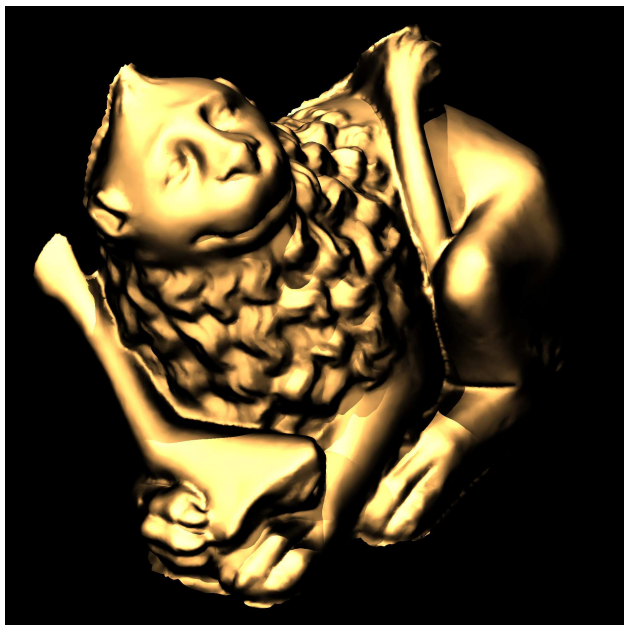


Figure 12: The lion statuette in opaque bulk gold only.

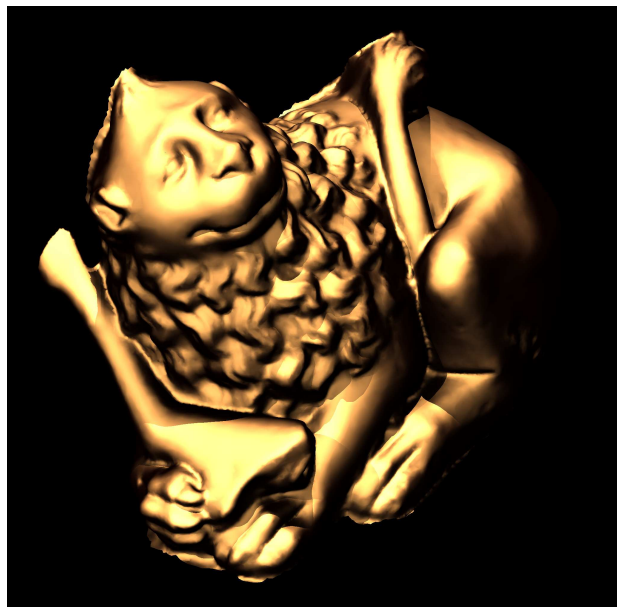


Figure 14: The lion statuette with gold leaf on a cinnabar bole. 5% holes and cracks.

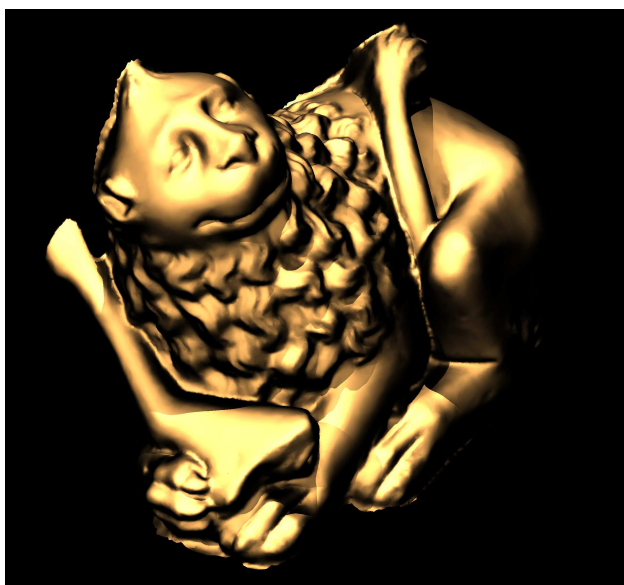


Figure 13: The lion statuette with gold leaf on a black bole. 5% holes and cracks.

model for rendering in spectral ray-tracing. The validation process to consider is not enough satisfying four quality search in rendering and will have to be improved. A comparison with the coloured materials used in substrate on wood also has to be continued. The virtual lighting conditions must be improved using the polarization of the reflected light accounting for the roughness of the metallic gold film. The statistical distribution of the holes and cracks has to be studied on many samples of gilded surfaces. For a more realistic visual appearance we have to implement the surface irregularities at different scales and starting from $50 \mu\text{m}$ spatial extension. Using optical images of real gilded surfaces, as we made, a displacement mapping technique enriched by the profilometric data, could give, at short viewing distances, some very convincing virtual images enough plausible for use in restoration proposal.

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