

Painted or not painted? Discovering color traces of ancient stones

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ABSTRACT

Although many ancient civilizations are known to have made use of polychromy on sculptures and in general on stone artifacts, today much of these colours went lost. For this reason, in the minds of a very large majority, the original stones have remained un-coloured until today. The small amount of these traces lead to a new approach for their characterization in order to limit sampling and hopefully, avoiding it. The non-invasive approach permits the examination of a very large number of artworks with a virtually limitless number of analytical acquisitions allowing to perform measurements *in situ*. Already during the measurement process, this approach leads to a fundamental exchange of views among scientists, archaeologist, conservators and art historians.

The application of protocols based on imaging techniques (i.e. UV Fluorescence, Visible Induced Luminescence-VIL) integrated with data obtained from single spot techniques such as X-Ray fluorescence (XRF), Fibre Optic Reflectance Spectroscopy (FORS) and Total Reflection Infrared Spectroscopy (TR FTIR), provides high-quality information. In this paper some examples of analyses conducted in different contexts from museums to archaeological sites will be presented. These analyses are included in a wider research project aimed to enlighten the use of colours on the sculptures in ancient time and to better define materials used in the past

KEYWORDS ancient stone, polychromy, integrated protocols, archeometry

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1. Introduction

In recent years, a strong interest about the residual polychromy on marble statuary emerged. Although many ancient civilizations used polychromy on stone sculptures and architectural elements, today only a few, almost invisible, traces of these colours survive. As a result of these losses, in the minds of a very large majority of people, sculpture and monuments are considered not to have been coloured since their creation.

Understanding ancient polychromy is indeed a crucial issue since the lack of attention to this theme leads to a significant misunderstanding of both the artwork itself and of the artistic culture it represents.

A correct reading of the original aspect of an artwork is often difficult due to the small amount and condition of the colour remainings. This is linked to the vicissitudes the sculptures underwent over the centuries, such as exposure to harsh environment or burial. In some cases, the polychromy did survive over the centuries, only to be extensively removed after hasty archaeological excavations, in order to reveal the Neoclassical white «pure form» of the sculpture.

The debate on the polychromy of ancient sculpture was already active in the nineteenth and early twentieth century, but only in 1982 it did a breakthrough in the studies with the research project initiated by von Graeve on the polychromy of ancient sculptures (Von Graeve 1985).

From then on, the issue of colour received a growing interest. In archaeology, a rising importance was given to

the role of colour in understanding ancient cultures. Furthermore, the technological development of non-invasive or micro-invasive tools supported these emerging archaeological ideas with archaeometric studies.

These new studies and collaborations were particularly evident starting from the travelling exhibition *Bunte Götter*, Munich (2003-2004); *I colori del bianco*, Rome (2004); *ClassiColor*, Copenhagen (2004). This exhibition was born by the collaboration among V. Brinkmann, J. Østergaard and P. Liverani (Brinkmann and Wünsche 2004), pioneering scholars in this field. This high-profile exhibition of painted Greek and Roman casts, carefully studied and reconstructed by experts from across Europe, attracted extensive media attention and turned out a renewed interest in sculptural polychromy in both popular and academic circles. This international initiative set the scene for a number of important projects, leading to a very noticeable increase in documentation and publications (Giachi et al. 2007, Liverani 2009, Liverani and Santamaria 2014).

2. Methodological approach

The archaeometric studies allow gathering information about the material composition and the state of conservation of polychrome marble artworks. The small amount of these coloured traces lead to a new approach for their characterization. In addition, since traces of polychromy are rare and fragile survivals, non-invasive methods are always preferable. In this way, the residues of polychromy remain intact for future generations.

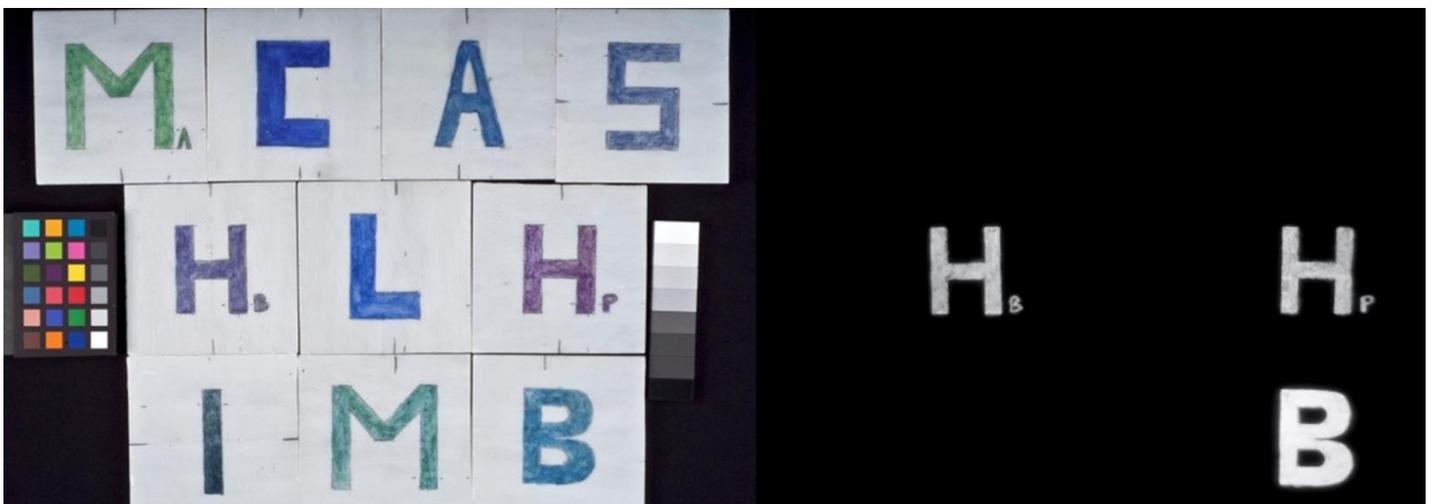


Fig. 1: Visible (left) and VIL (right) images of marble mock-ups painted with different pigments (tempera): M=malachite; C=Cobalt Blue; .A=Azurite; S=Smalt; H_B= Han Blue; L=Lapis lazuli; H_P= Han Purple; I=Indigo; M=Maya Blue; B= Egyptian Blue.

A non-invasive approach permits the examination of a very large number of artworks with a virtually limitless number of analytical acquisitions allowing to perform measurements *in situ*. Already during the measurement process, this approach leads to a fundamental exchange of views among scientists, archaeologists, conservators and art historians.

The non-invasive scientific protocol proposed, is characterized by the combination of complementary analytical techniques. The procedure started with a preliminary documentation of the surfaces by means of multiband imaging. This survey is based on photographic techniques using different radiations (Fischer and Kakoulli 2006, Dyer and Sotiropoulou 2017, Dyer et al. 2013, Cosentino 2014). UV fluorescence (UVf) (or more correctly, Ultraviolet-induced visible luminescence (UVL)) was used to spatially characterize the presence of organic and inorganic materials. Visible Induced infrared Luminescence (VIL) was used to locate and identify the blue pigment Egyptian blue. VIL is a photographic technique developed at the British Museum in 2009 that can detect the Egyptian blue which is a calcium-copper based pigment ($\text{CaCuSi}_4\text{O}_{10}$) (Accorsi et al. 2009, Verri 2009).

Egyptian blue is often preserved in extremely small quantities in the porous surfaces of ancient objects, which makes it hard – if not impossible – to identify the pigment even with a microscope. The pigment has, however, the unique property of absorbing visible light and emitting it as infrared radiation. The luminescence emitted by the pigment grains can be recorded with an infrared camera. The technique thus exploits the powerful emission identifying single particles of Egyptian blue, which would be otherwise undetectable (Verri 2010). The luminescence phenomenon is illustrated in Figure 1 where the image in visible light and the VIL image of marble mock-ups painted with different blue/green pigments are compared. The white glow represents

Egyptian blue (B) and two Chinese pigments (Han Blue and Han Purple) while the others pigments almost disappeared.

The efficacy of a multi-analytical approach is evident in Figure 2 in which an area of a decoration on a sarcophagus with tiny trace of polychromy is investigated under different wavelengths and filters combinations (Iannaccone et al. 2015).

Addressed by the imaging results, analyses by X-Ray Fluorescence Spectroscopy (XRF) (Karidas et al. 2006, Shugar 2012), UV-VIS Reflectance Spectroscopy (FORS) (Bacci 2000) and Total Reflection Infrared Spectroscopy (TR-FTIR) (Miliani et al. 2012) are performed in order to gain molecular and elemental information on a wide range of inorganic and organic painting materials, including most pigments, colorants and binders (Iannaccone et al. 2015, Liverani et al. 2017).

Today, despite important results achieved within the last few years, research on ancient polychromy is still at an early stage and shows some obvious limitations pertaining to the transferability of methodologies or findings (Gasanova 2018). Some interdisciplinary teams already integrate analytical and archaeological competences. One example is the work done at the Ny Carlsberg Glyptotek in Copenhagen, within the framework of the Tracking Colour project (Østergaard 2010). In the published reports (Ny Carlsberg Glyptotek 2013), a multidisciplinary approach to study a large group of materials is highlighted.

The same approach, a collaboration with archaeometric competencies and the archaeological ones, drove our researches in the last years (Iannaccone et al. 2015, Liverani et al. 2017, Noferi 2017). In this paper some examples of findings in different contexts from museums to archaeological sites will be presented.



Fig. 2: Example of application of imaging technique on a polychrome marble sarcophagus. a) Visible image; b) UV image where the fluorescence of red lake is visible; c) VIL image where Egyptian blue particles (not visible at naked eye) are instead clearly visible.

2.1 Technical information

Multiband imaging (UV and VIL)

For the investigation two camera were used: a Canon EOS 7D with a resolution of 18 Megapixel and a modified Canon EOS 400D with a resolution of 10.10 Megapixel. Both cameras mounted a Canon EFS 18-135 mm f/3,5-5,6 IS lens with different filters on varying of every photographic technique applied. Different filters were applied also on the two flashes Quantum Qflash T5dR thus providing the proper radiation.

Fiber Optics Reflectance Spectroscopy (FORS)

FORS spectra were acquired in the range 350-900 nm using an Ocean Optics (mod. USB2000) instrument, equipped with optic fibers and a tungsten lamp as source. All the measurements were performed with 2x45°/0° configuration, allowing to work in diffuse reflectance by collecting the light scattered at 45° with respect to the incident light (avoiding specular reflected light) from an area of 2 mm in diameter. Each acquired spectrum is the average of 30 acquisitions. A Spectralon® tag was used as reference.

Fourier Transform Total Reflection Infrared Spectroscopy (TR FT-IR)

Infrared measurements were carried out by means of a portable Bruker Optics ALPHA FT-IR Spectrometer equipped with SiC Global source and a DTGS detector.

All spectra were acquired in total reflection mode, collecting 128 scans, with a resolution of 4 cm⁻¹ on the 7500-375 cm⁻¹ range and a measuring spot of 5 mm in diameter. The collected IR spectra were processed using OPUS 7.0.122 software.

X-ray fluorescence (XRF)

For X-ray fluorescence measurements, a portable XRF spectrometer (Bruker, Tracer III SD) with micro X-ray tube with rhodium anode was used. The analysis was performed in the following conditions: 40 keV - 12 µA. The irradiated area was about 12 mm². The spectrometer was equipped with an SDD detector (FWHM < 145 eV at 100.000 cps) cooled with a Peltier cell. Acquisition time for each spectrum was 60 s.

3.Examples of colour traces studies

3.1. Catacombs

The first case-study here presented is a sarcophagus with strigilated lateral parts and a central scene depicting two figures (a male and a female and several other objects and animals). It is preserved in the Catacomb of St. Pamphilus, on the *Via Salaria Vetus*, at the deepest level.

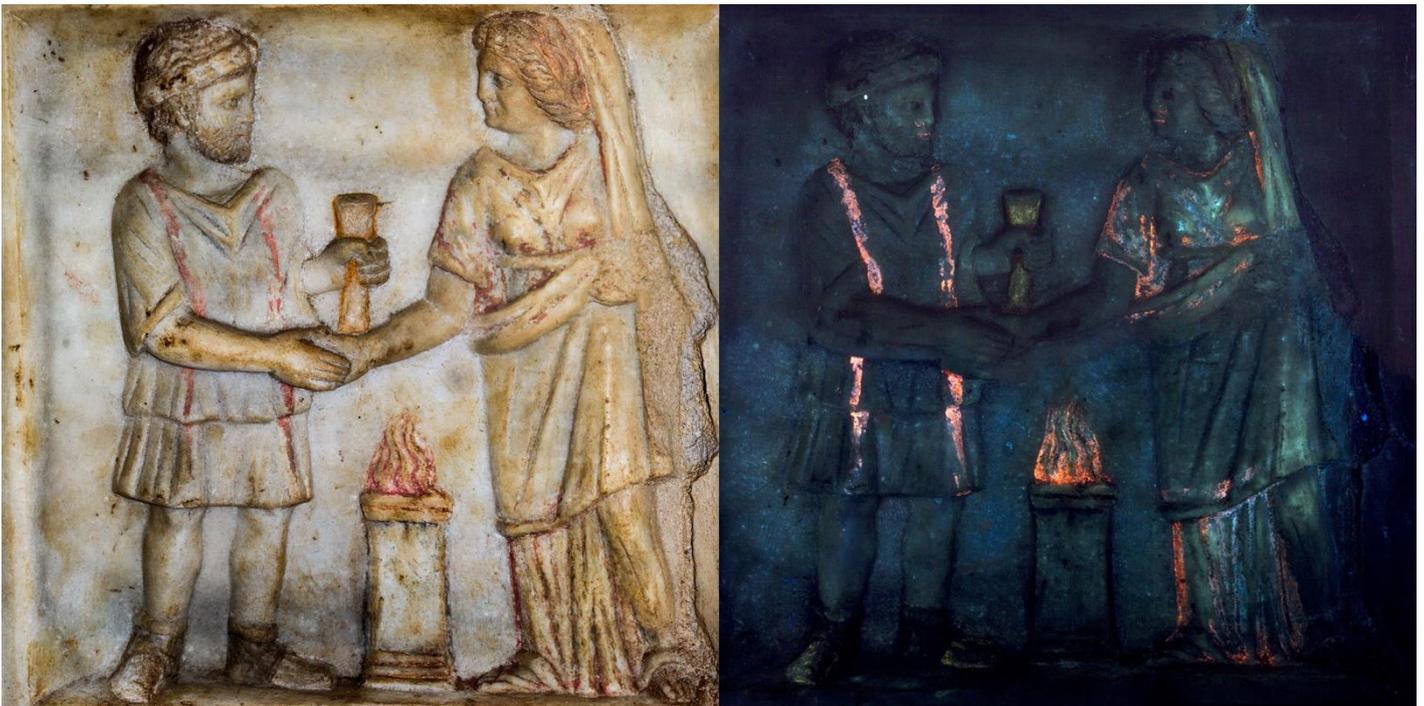


Fig. 3: Visible (left) and UV fluorescence (right) images of red lake on *Olimpus Antistianus* and *Irene*.

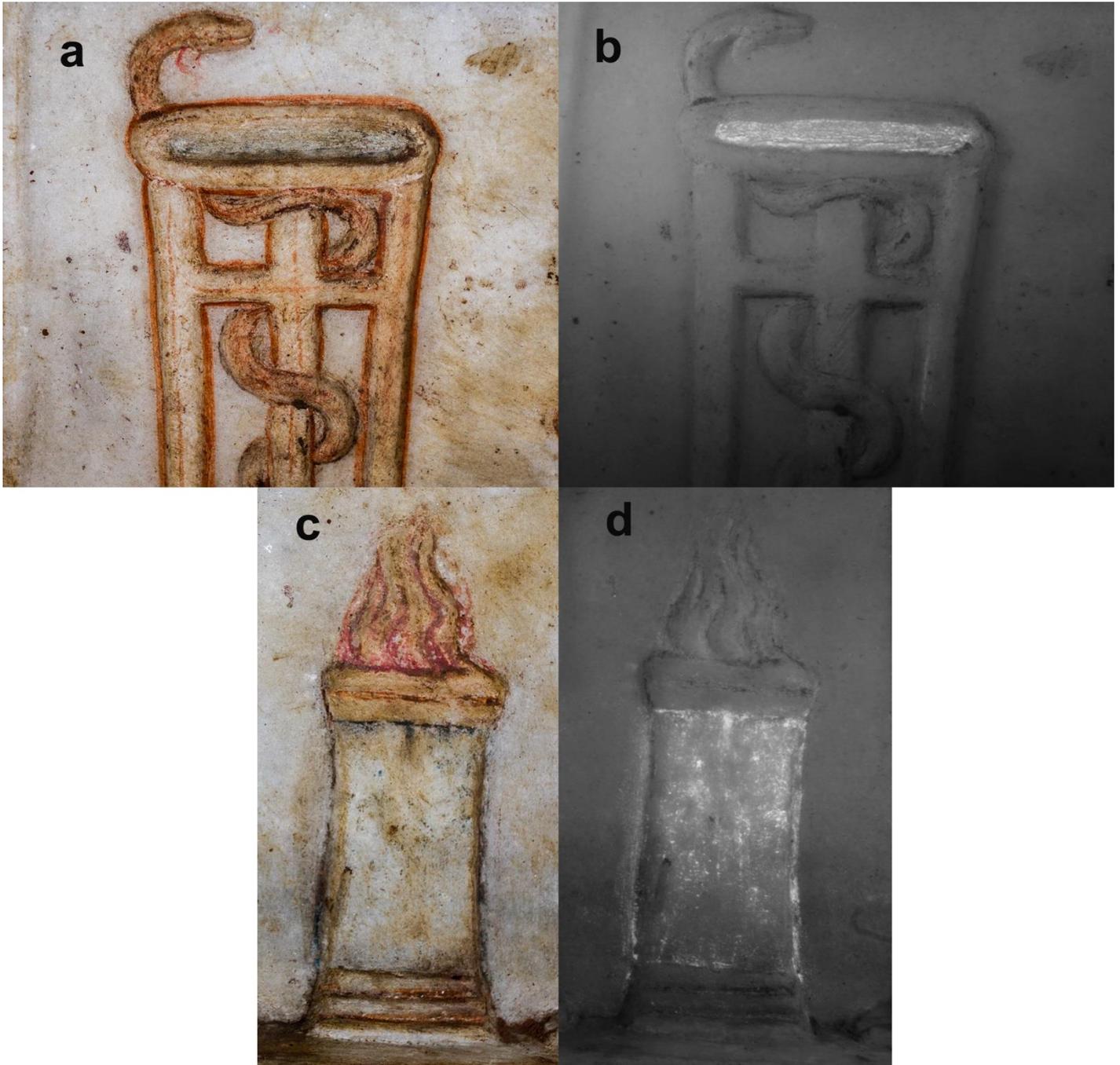


Fig. 4: Visible (a,c) and VIL (b,d) images of some details of the Antistianus sarcophagus

In the slab showing the *dextrarum iunctio* between Olympus Antistianus and his wife Octavia Irene, the surface revealed traces of red and blue colours, slightly visible to the naked eye, especially on the robes and the altar. All the red traces showed a characteristic ultraviolet pink-orange fluorescence (Fig. 3) that it is usually associated with red lake pigments such as madder lake enabling to hypothesize the use of a red lake for creating the red details.

Analyses using the VIL technique revealed the presence of Egyptian blue in some specific part of the bas relief such as the basin or the pillar of the flame (Fig. 4).

Data acquired with other spot techniques (XRF, FORS) confirmed the findings and the hypothesis drawn by observing UVf (data not shown) and VIL images. In this case, micro-climatic conditions of the catacombs are suitable for the preservation of colours on stone.

Temperature and humidity are constant and the lighting is limited to the duration of either visits or inspections. These conditions facilitate the conservation of polychromy, allowing deep investigations that can highlight important details about the technique used by the old master painters.

3.2. Outdoor and indoor statuary

Etruscan Gens Statlane's sarcophagi, are part of the Florence National Archaeological Museum collection, currently preserved in the courtyard of Villa Corsini, located close to Sesto Fiorentino. These sarcophagi are dated back to the first half of the third century BC.

The group consists of ten sarcophagi. Among all the sarcophagi, only the VIL survey performed on sarcophagus of Vel Statlane (Fig. 5) highlighted an interesting residue of Egyptian blue on the lower part otherwise not visible at naked eye.

Conversely, the sarcophagus belonging to Ramtha Ziltna (Fig.6), is the only one, among those analysed, showing the presence of red lake. The characteristic red/pinkish fluorescence, under UV light, appears on the belt and the ribbon of the woman portrayed on the lid. FORS spectra acquired on this area confirmed this hypothesis.



Fig. 5: Sarcophagus of Vel Statlane, son of Sethi, 275-250 BC, Villa Corsini, Florence (left) and VIL image of a detail of the lower slab (right).

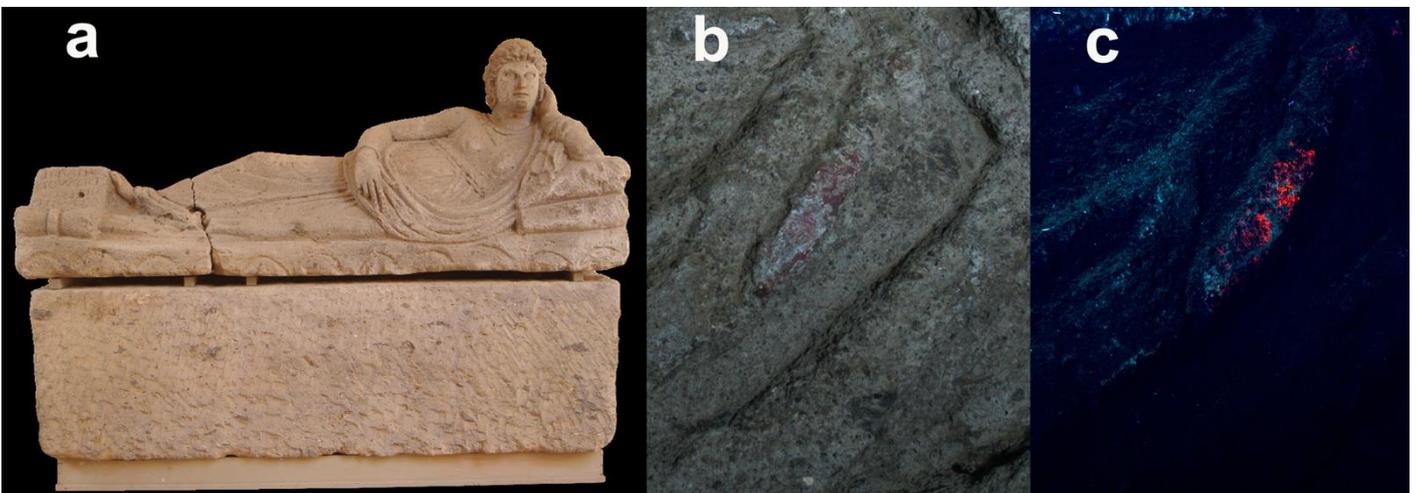


Fig. 6: Tomb II, female sarcophagus of Ramtha Ziltna, 260-50 BC, Villa Corsini, Florence (a), visible (b) and UV fluorescence (c) images of the detail of the ribbon.



Fig. 7: Headless Cuirassed Emperor (a) at the Archaeological Museum of Grosseto, macro image of residual gold (b) and VIL detail of the cuirass (c).

On other sarcophagi belonging to this group also traces of red and yellow ochres were identified (data not shown). The rare traces, almost invisible, discovered during the study did not allow us to establish with

certainly the extent of polychromy, but for sure the sarcophagi were painted. This is also confirmed by the documentation of the excavation and by the observations

of scholars shortly after the excavation. The sarcophagi were always described as polychrome artifacts.

The poor conservation of polychromy may be correlated to the lithotype used (Nenfro stone, Tuff), that is coarse and prone to disaggregation but also to the stressful conservative history. Indeed, after the excavation, at the beginning of 20th century, the sarcophagi were displayed in the courtyard of the museum, exposed to light and rain for more than 50 years, where in 1966 they also suffered the dreadful event of the flooding in Florence.

3.3. Excavated statuary

An interesting example of residual polychromy is represented by the Headless Cuirassed Emperor (Fig. 7a), belonging to early Imperial cycle from the *Augusteum* of Rusellae (Roselle, Grosseto) exposed in the Archaeological Museum of Grosseto. Tiny traces of gold have been discovered on the drapery (Fig. 7b), and traces of Egyptian Blue survived on the cuirass (Fig. 7c).

Also in this case few traces of colour/decoration were identified but they were enough to confirm the practice of painted and coloured statues. Even in this case the poor conservation of polychromy, apart from other reasons, is surely closely related to conservative history. From documents found during the study it emerged that after the excavation (in the 1950s) the statues were cleaned by immersing them in tanks with sodium hypochlorite and "heavily brushed" to remove the excavation earth.

4. Virtual reconstruction of the ancient colour

Digital reconstruction of ancient polychromy is a relatively recent issue in the history of archaeological and architectural heritage documentation. It emerged as a result of new interest in the experimental archaeology and the technological development of computer graphic tools.

The standard research activity has been recently supported by the development of experimental approaches, often based on digital technologies to propose and assess reconstruction hypotheses. Those hypothetical reconstructions of the original colours and decorations, previously exemplified on physical replicas of objects, are now moving to the digital media. They are usually reproduced on digital photorealistic three-dimensional models, obtained with scanning technologies (Siotto et al. 2014). The colour reconstructions, based on the results of scientific analysis and archaeological data, allow the visualization of the original appearance of the

artwork helping scholars to understand how and why colour was used to decorate or finish the artworks.

Anyway, the reconstruction of the original polychromy is not yet a consolidated subject of research; a lot of work still has to be done to improve our knowledge of the methods and techniques of colour application on polychrome artworks (Østergaard 2010).

In addition, the virtual reconstruction becomes more difficult in the context of ancient polychromy, because just tiny and deteriorated samples are usually found.

Today, MeshLab software (MeshLab 2019) was used to support the polychrome reconstruction stage and Blender (Blender Python API 2019) (or, rather, a combination of MeshLab and Blender) was used to achieve a more sophisticated visual presentation of the current and reconstruction ancient colour (Siotto et al. 2015).

Previous projects tried to realistically simulate the original colour of the works of art. An interesting example was the one supervised by Prof. Paolo Liverani, which returned a 3D model of the Augusto di Prima Porta at Vatican Museum, complete with its decorative apparatus (Liverani 2004). In 2002, the Stone Restoration Laboratory of the Vatican Museums started a careful and painstaking cleaning which has brought back to light many traces of colour that were no longer visible. To understand the nature and the composition of the pigments, they were subjected to a scientific examination by analytical techniques. The results of these investigations showed that colours were applied on the clothing, on details of the armour, on the hair and on details of the eyes but not on the skin or on the ground of the armour.

On the basis of these evidences, a complete reconstruction of the colours was prepared on a plaster cast. The surviving traces are sufficient to permit the colours to be reconstructed over most of the surface. The missing parts were supplemented in a hypothetical manner based on the logic of the use of colours on the statue and on comparisons with roughly contemporary portraits (Liverani 2011).

5. Conclusions

The detailed knowledge of an artwork or archaeological artefact, in terms of its composition, is a prerequisite condition for any research in art history or archaeology as well as for any conservation-restoration procedure. The scientific approach here presented represents a strategic tool for achieving a complete awareness of the residual polychromy on ancient statuary; it will open up new understanding of original polychromy, supporting more

correct and conscious restoration procedures (e.g. in the case of the archaeological excavations or programmatic operations in the museum collections). The aim is to develop a broader awareness of residual polychromy to be shared with the research groups active in this field on the national and international scene, with the museums and institutions involved in conservation of cultural heritage and for educational purposes with non-specialist public.

The technical examination is based on the use of several non-invasive methods. In this way, a very large number of artworks with a virtually limitless number of analytical acquisitions can be analysed. This approach leads also both to the reduction of the sampling activity and to a fundamental exchange of views among scientists, archaeologist, conservators and art historians already during the measurement process.

One still open issue is represented by the assessment of the reliability of the digital colour reconstructions. Until now it has not been possible to create realistic renderings that take into account the effect of colour with the material (marble or other supports) and light.

Further improvement could be dedicated to testing the actual system to assess the effectiveness and limitations to recreate a hypothesis of the original colour (on the digital reconstruction).

Very often the extent of the surviving pigments/colour is too small to allow a satisfactory reconstruction of original appearance. The latter is a combination of several parameters, such as pigments but also binders, thickness, preparation layers, etc.) that are not always known. So the reconstruction is something very risky that can easily turn into a modern reinterpretation.

6. Conflict of interest declaration

All authors states that there is no conflict of interest.

7. Funding source declaration

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9. Short biography of the authors

Susanna Bracci - She is Senior Researcher at ICVBC-CNR. She is leading the ICVBC Mobile laboratory for the in-situ diagnostics of works of art including paintings, frescoes and glasses (mosaics and stained glass windows). In this framework special attention is devoted to the color traces on statues and architecture.

Donata Magrini - She is a Conservation Scientist at ICVBC-CNR. Her research activity is mainly aimed at investigating methodologies for diagnosis and monitoring the state of conservation of cultural heritage, with special attention to paintings, frescoes and stones. In this context, she is involved in the application of non-invasive techniques (UV-Vis-NIR spectroscopy, XRF, FT-IR and imaging) to study residuals of original polychromy on stone.

Giovanni Bartolozzi - He is a Conservation Scientist at IFAC-CNR. His research activity is focused on the diagnostic for Cultural Heritage (wall paintings, easel and canvas paintings, contemporary artworks). He is expert in both non invasive and invasive spectroscopic techniques (FORS, UV-Vis-NIR, FT-IR).

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