

MODERN NON-DESTRUCTIVE PHYSICAL METHODS FOR PAINTINGS TESTING AND EVALUATION

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ABSTRACT

There are several rapidly developing non-destructive physical methods that are increasingly used for the analysis and assessment of art objects. These technologies are posed to bring about drastic changes in the attribution and inventory characterization procedures carried out by museums, private collections, art dealers, and art community as a whole. Certainly today nondestructive evaluation (NDE) of art objects is becoming more and more sophisticated. The applications of these methods include but not limited to deterioration detection, authentication, revealing forgeries and fakes, and attribution. There are numerous physical techniques that are currently adopted for various tasks related to NDE in the art world, many of which have been specifically developed for particular applications of paintings evaluation. While some of these methods are well-known, others remain rather obscure. This article is an attempt to review a few physical techniques that could be successfully used for the conservation and authentication of paintings. The methods described here include multi-spectral illumination methods, thermographic techniques, scanning acoustic imaging, air-coupled ultrasound, Raman spectroscopy and imaging, and Mass-spectroscopy.

INTRODUCTION

It is perhaps appropriate at the outset of this review to describe the external factors which affect the condition of a painting. Certainly, the influence of temperature changes, exposure to light, inappropriate handling, bacterial contamination insect infestation, can contribute to changes in appearance and physical condition of a painting. Further, with such aging paint layers get more brittle leading to cracks or even the paint detachment from support. If left untreated, with time the painting can become unrecognizable or can even be irreversibly damaged. In order to properly preserve or restore a painting it is particularly important to know the physical/chemical mechanisms of degradation effects. The most common are:

- *Darkening and change of color* – There are several possible reasons for color to change. The painting accumulates deposits of common room dust, soil particles, and particles of organic origin on its surface. Usually such deposits can be easily removed with wet cotton. Other mechanisms include the oxidization of varnish and/or various chemical reactions that take place in the paint layers.
- *Craquelure* – a network of fine cracks or crackles on the surface of a painting, caused chiefly by shrinkage of paint film or varnish, but could also be a result of weakening of the ground and/or support.
- *Layers separation* – Delaminations between layers of paint, between the paint and the ground, or possibly between the ground and the support. This phenomenon can be attributed to that fact that different paints have dissimilar expansion properties. Also, the ground and paint layers have different stiffness [1, 2]. The most common reason for layers separations to occur is due to changes in the material stiffness with age – the paint layers become more rigid, while the ground loses stiffness. All of these factors contribute to accumulating stresses and consequent delaminations.
- *Biodeterioration* – regions of some paintings contain biological components; they can be source of food for insects, larvae, or even small rodents. For example, woodworm or a larvae of certain wood-boring beetles (for example Ambrosia beetles (Platypodidae, Scolytidae), Common furniture beetle (*Anobium punctatum*), Death watch beetle

(*Xestobium rufovillosum*), House longhorn beetle (*Hylotrupes bajulus*), Powder post beetle (*Lyctus brunneus*), or a Weevils (*Pentarthrum huttoni*, *Euophryum confine*) often bores wood stretchers, wood supports, or frames, weakening them and creating a risk of severe layers separation.

- *Biodegradation* – paintings are often affected by biodecay. Fungi and bacteria cause severe damage through mechanical processes from growth into the painting and its grounding and through their metabolism. This may result in the embrittlement of the ground, layers separation, and/or the deterioration of the canvas or wooden support.

While some of these deteriorations are easily observable, others become visible only when the damage is extreme. For example, if layers separation is allowed to cover the majority of the painting area, the restoration process is almost impossible, requiring a large amount of time and labor. Thus the use of advanced subsurface non-destructive methods can prove extremely beneficial providing information on often invisible damage. The information can be used to preserve the painting by arresting or reversing the damage to avoid lengthy and sometime risky restoration procedures.

Forgery is another major issue of the art world. Needless to say, ancient art, historical materials, and old masterpieces are treasured for their cultural and collection values. As such, these pieces of art have changed hands numerous times during the course of their life. Some experts say that the problem of fakery is as old as the art market itself [3]. Naturally, then, it is of great interest to the art community to discern between real and imitation artifacts. Thus, methods for revealing the forgeries have continued to develop over time. Today, the majority of museums and auction organizations employ numerous art experts having great experience in this field. Unfortunately, the quantity and quality of forgery techniques are improving, resulting in imitation works of art that are increasingly able to fool even the most experience expert.

It is for this reason that there is an increasing interest in the development of analytical methods and tools that have sufficient sensitivity and resolution to identify inauthentic artwork. These tools and techniques will serve as quantitative methods upon which decisions regarding the authenticity of a painting could be based.

In this paper we will limit ourselves to techniques that are based on non destructive electromagnetic irradiation ranging from infrared (IR) to ultraviolet (UV), X-rays, and acoustic irradiation. Specifically, these methods are: near-infrared technique (NIR), various X-ray techniques, UV analysis and "black light" techniques, thermography, various acoustic techniques, Raman spectroscopy and imaging, polarization optical microscopy, optical coherence tomography, and radioactive elementary analyses.

The spectroscopic examination of artworks is of high importance for conservators, art-historians and keepers of private or public museum collections. These investigations reveal information, which is of general (art- historical interest: the knowledge of the artistic materials that were used and were available at a certain period in a particular region and the dissemination of new methods provide information about the interactions between distinct cultures and about trade routes. The knowledge on the artists' materials that were available in particular regions and periods can help in dating artefacts.

To study painting's features which are hidden under or within layers of paint, under dust and darkened varnish, methods such as "black light", near-infrared, X-ray, or acoustic techniques

are preferable. These methods, well known and widely adopted for the NDE of art [4], allow "penetration" through the covering layers, revealing features hidden below. With respect to delaminations, thermographic methods, as well as contact and non contact ultrasound and scanning acoustic microscopy can be utilized. It should be noted, however, that although acoustic microscopy and various air-coupled techniques have been referred to in publications as viable methods for finding layers separations in wooden paintings [5, 6, 37], their use in the non-destructive evaluation of paintings remain relatively novel. Both of these techniques will be described in detail in subsequent sections of this paper.

In order to study and identify different pigments, it is necessary to better understand the molecular structure of the paint. Some paints have complex molecules in their pigment-binder structure which can be identified using Raman spectrographic techniques [7-9]. Optical microscopy enhanced with polarizing techniques is also able to differentiate the crystal structure of pigments, revealing different kinds of non-organic pigments in paints [10, 11].

In cases where the structure of the layers is of interest, optical coherence tomography is a strong candidate [12-14]. This technique uses a vertical scanning interferometer-based stage with a light source having an extremely short coherence length. As the length of coherence is extremely short, constructive interference occurs only when the distance to the reflective surface is equal to the length of the reference path. Spatial scanning while changing the reference path length allows one to collect a "cross-section" of paint layers along the desired path.

In our Institute we deal with the majority of those techniques: visual, near-infrared, thermographic, acoustics, X-ray and Raman techniques.

VISUAL EXAMINATION

Visual examination is the simplest and most natural way of performing nondestructive evaluation. It is, however, a very important component of any analysis. For paintings, visual examination includes naked-eye observations with or without the use of a microscope. Naked-eye visual examinations can provide information pertaining to late stages of deterioration such as transparency of the varnish, color balance, and cracks. In effort to acquire more precise and detailed results, restorers typically use optical microscopes with relatively moderate magnifications. Unfortunately, this method is usually restricted to testing surface condition only since the majority of paints are opaque for the visible light spectrum.

On the other hand, visual examination is used by many experts to check the style of a painting. For example, the shapes of the brush strokes can provide information about the period of the painting and/or the actual author [15]. Such examinations, however, require vast amounts of experience. Some art researchers introduced computer recognition software into this field by combining optical scanning and various image processing techniques [15, 16]. Inspection by means of "black light" also can yield valuable information about a painting. For example, on a painting alleged to be Ruben's "Roman Charity", "black light" illumination revealed a vertical line which did not pass through the entire painting. Instead, it occurred only in those regions having no painted foreground object. Further study showed that the *craquelure* had a different structure along that line all over the painting. This suggested that the painting had been folded sometime ago and that the most severely damaged regions had been subsequently restored (Fig.1).

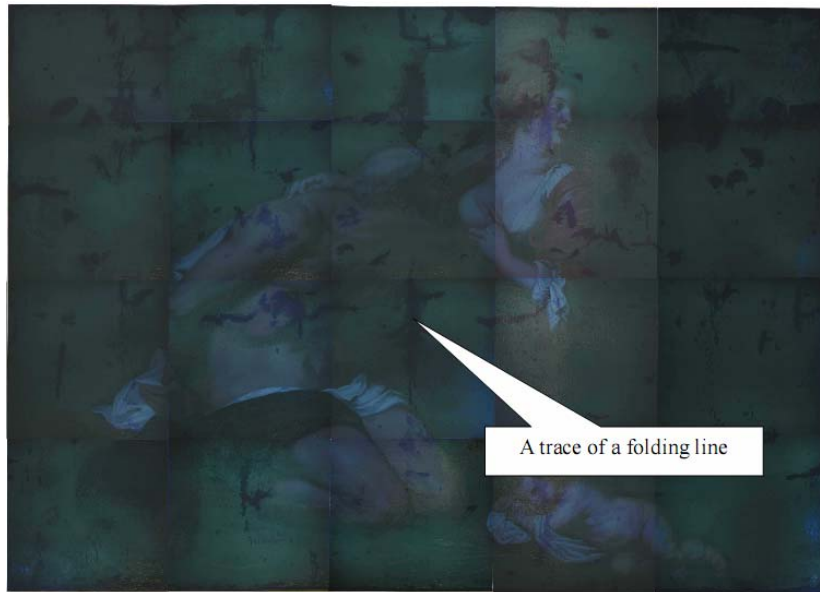


Fig.1. Visual inspection of a painting in near-ultraviolet fluorescence (a mosaic-built image).

NEAR-INFRARED (NIR) RADIOGRAPHY

NIR radiography method is excellent for authentication of painting in that it is able to reveal hidden underpaintings. Near-infrared (0.7-2.5 μm) band is able to penetrate the pigments more readily than visible light. In fact, it has been shown that some pigments like ochre, lead white, vermilion, exhibit minimal absorption for wavelengths around 2.0 μm [17]. This allows one to observe the distribution of reflected and absorbed near-infrared light thus revealing covered layers, allowing one to find sketches, outlines and versions done by an artist on the ground. Highly reflective sketching materials such as graphite are easily recognizable.

This method was described by J.van Asperen de Boer at the University of Amsterdam in the middle of the 1960s [17, 18]. On the basis of that method, Kubelka-Munk analyzed the optical properties of paint films. The early systems used NIR vidicon cameras or NIR-sensitive films with various additives to enhance their sensitivity [17]. According to several authors [17, 19] the optimal NIR detector should have peak sensitivity close to 2.0 μm . That makes PbS one of the most suitable detectors for the purpose of NIR examination. After 2.0 μm the absorption in most of paints increases due to O-H and C-H vibrations in the pigment molecules [18].

Recently, Si-based CCD-matrices have come into use [19]. It is well known that standard CCDs used for digital photography detect not only visible band, but also part of the near ultraviolet and near infrared band (usually up to approximately 1.0 μm). Such modern digital cameras can be effectively used for the detection of underpaintings as well [3, 20]. For this reason we have begun to use it in our own art research. For NIR examinations we currently use Fujitsu FinePix S3 Pro UVIR forensics camera with a PECA filter 904 (Wratten 87B, IR transmission band from 900 nm). Using this camera we successfully detected hidden sketches (Fig.2). The report on this work is included in another presentation at this Conference.

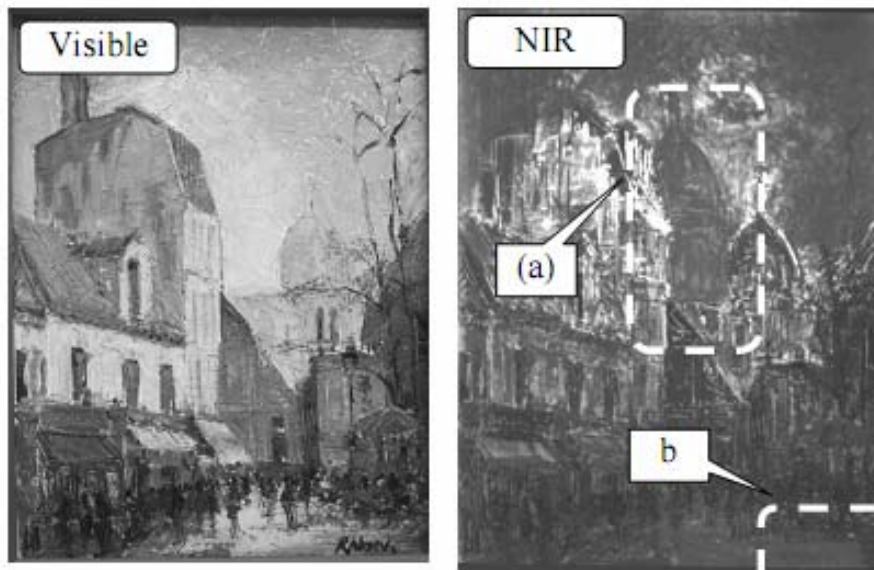


Fig.2. NIR examination of a painting: (a) Hidden chapel, (b) Faked signature (a painted region is visible).

A "mosaic" approach can also be used as an alternate technique [21], constructing the entire NIR-image piece by piece from several images made by an NIR-operating camera. This method prevents large scale optical image distortions.

Returning to the previously mentioned "Roman Charity", NIR reveals more details that were hidden under the darkened varnish layers. These include the same line which was restored after the painting was folded (Fig.3).



Fig.3. NIR examination of "Roman Charity":
 (a) Restored region of the fold line, (b) A cloak, or a shawl, visible in NIR only.

THERMOGRAPHY

Thermography uses middle (3-5 μm) or far (8-14 μm) infrared illumination (or a "heat pulse") for nondestructive diagnostics. Here the inspection is called *active* thermography—in contrast to *passive* thermography where the heat emitted by the sample itself. Thermography is a widely used NDE method for the examination of the internal structure of various materials including metal joints [22-24], multilayered composite plates [25], plastics, and paper [26]. All thermographic methods are principally based on extracting data from the heat dissipation dynamics after a sample was irradiated by the external source. The dynamics is studied by means of a time sequence of images captured by a thermal camera. The keystone of the method is that the cooling rate is faster for sound (good) areas than for defective (damaged)

areas of the painting. This dynamic difference (i.e., the thermal contrast) provides information about location and shape of defects.

There are three different kinds of active thermographic inspection – pulsed thermography (PT), lock-in thermography (LT) and pulsed-phase thermography (PPT) [27-29].

In PT, a pulse of heat (or cold) is applied to the sample. The shape and duration of the pulse can be adjusted for certain conditions of the experiment or for a particular sample. When the surface is heated uniformly, all points cool at the same rate only in the absence of defects. If, on the other hand, the heat wave encounters defects having lower thermal conductivity, the rate of cooling at that point changes. This information can be detected with a thermal imager. This is the most widespread method of thermal testing and is often employed to find hollows in wood and delaminations in paintings.

Lock-In thermography uses harmonic heat waves rather than heat pulses. It permits the discovery of inclusions and delaminations at different depths in the bulk of materials with low thermal conductivity. Due to this fact, these experiments generally require longer periods of time to complete. This method is, however, is good for the examination of samples such as concrete, wood, marble, pottery, etc [30].

PPT combines both of the above described examination techniques. It is well known that a short pulse can be Fourier-transformed into a series of harmonic waves. Thus, a short pulse can be treated as a number of harmonics acting at the same time. Converting the resulting temperature dynamics, one can obtain results similar to those of LT at different frequencies. Following its development in the 1990s by X.Maldague [31], this method has been widely adopted for metal [22, 23], Plexiglas [32], composites and layered materials [25]. A recent modification of PPT uses wavelet-transformation rather than Fourier-transformation [33]. As suggested earlier, a painting can be described as a multilayered structure composed of several layers having different properties. In other words, it can be treated as a composite structure and, as such, can be tested with thermography. This allows one to detect both delaminations and restored areas [34]. This technique can also provide a complimentary method for detecting underpaintings or damages in ceramics and architectural monuments [35, 36].

Pulsed phase thermography is a good method for uncovering the genuine artist's signature hidden under layers of paper and paint as shown in Figure 4.



Fig.4. Revealing of hidden features of a painting.

Unfortunately, despite its great utility, the active thermography remains a rather exotic and rare NDE method for examination and evaluation of paintings.

SCANNING ACOUSTIC MICROSCOPY (SAM)

In recent years Scanning Acoustic Microscopy has reached spatial resolutions comparable to that of optic microscopes [37]. Since the contrast mechanisms depend only on the elastic

properties of the materials under examinations, these instruments provide a valuable method for materials characterization in bulk or subsurface zones that are not obtainable in any other manner. Acoustic microscope images are commonly performed for a variety of biomedical tissues, microelectronic circuitry, and for residual stress/strain imaging of a variety of materials [37]. SAM utilizes sound pulse-echo or through-transmission methods to detect cavities, hollows and/or inclusions. The reflection scheme is more common in that it allows on to study multilayer structures; transmission modes yield integrated data with less information on layers configuration. Fig.5 demonstrates the capabilities of this technique to measure the shape of various objects. The Tessonics[®] acoustic microscope has been used for this purpose. This particular device allows three-dimensional data to be stored and subsequently processed. In the examples shown, an algorithm based upon the use of Hilbert transforms was used. This algorithm enhances the temporal resolution of each A-scan by finding the envelope of the signal.

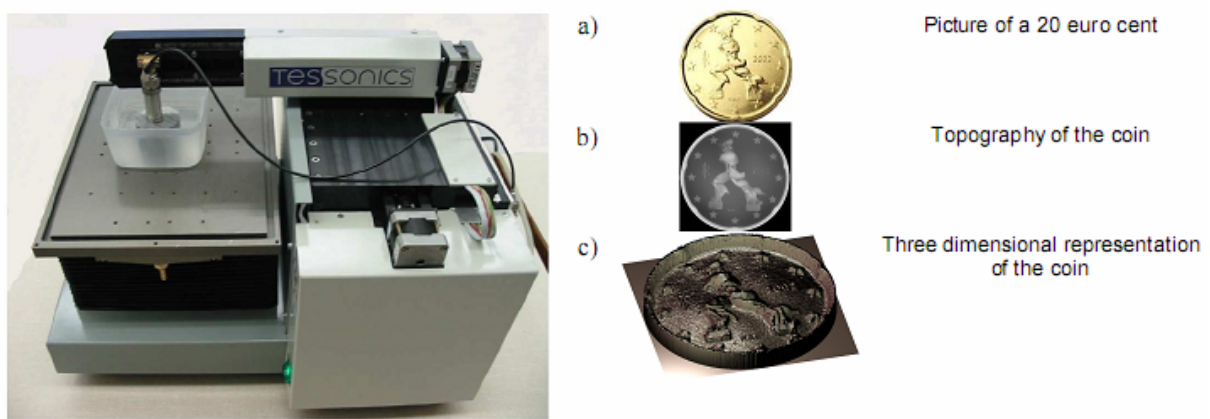
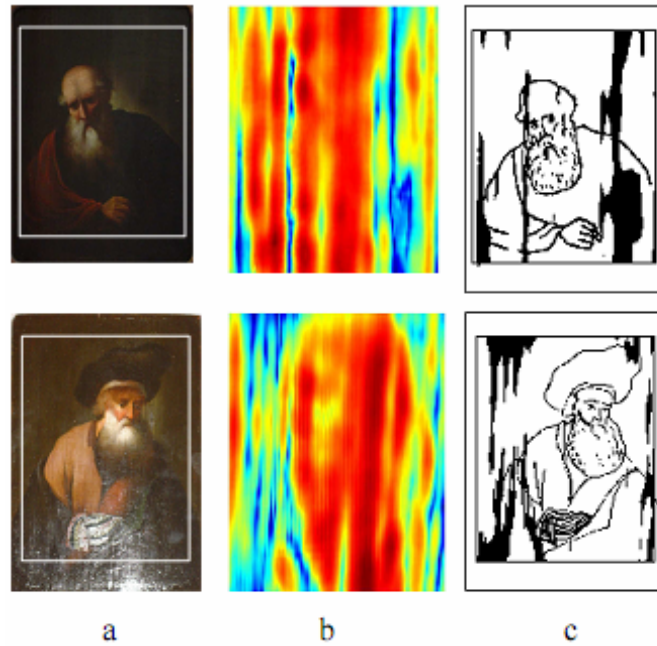


Fig 5. Tessonics[®] acoustic microscope (Frequency 10-400 MHz, Resolution: from 500 μm (10 MHz) up to 10 μm (400 MHz). a - is a visual picture b,c are scanning results of a 20 euro cent coin using 25 MHz focused transducer.

In Fig.5, the scanning results of a 0.20 euro coin are illustrated. A 25 MHz focused transducer was used. In Fig.5.b, the out of plane displacements are shown using a 2D representation form, whilst in Fig.5.c it is possible to see the corresponding 3D image [5].

AIR-COUPLED ULTRASOUND

Air-coupled detection systems, having been optimized to monitor the mechanical tape laying process for the manufacture of various multilayer composite structures, are very gentle in nature. As such, we consider it to be a very promising technique for the purposes of the Conference. In order to test wood-panel paintings, our team modified an NCA1000-2E (Ultran Laboratories) air-coupled instrument with a scanning option [5, 6]. The results revealed the position of delaminations as well as regions of wooden cracks with high precision, both of which are very important to restorers. In Fig.6, the scanning results for the XVI century wood panel painting are illustrated for the air-coupled transducers in the reflection mode. In Fig.6.a, a picture of the examined painting is shown together with a white rectangle which indicates the region that was examined using the acoustic microscope. In order to see the ultrasound reflection from the delaminated region (located at about 1.5 mm under the painting surface), a low frequency focused transducer was used (3.5 MHz). It should be noted that the high attenuation of the wood does not permit the use of higher frequencies. In Fig.6.b, the C-scan at a depth of about 1.5 mm is shown; the detached region was clearly detected. In Fig.6.c, a binary representation of the complete data is illustrated.



*Fig. 6 Non contact air-coupled scan in single-sided configuration.
Binary images with highlighted the delaminated regions (White - sound zones; black - detached zones).
The scanned region is that one inside the rectangle.*

For paintings examination purposes, leaky guided waves are a promising variant. This scheme uses oblique incident waves to generate guided waves in the upper layers of the sample. While propagating along the surface, this wave creates a leaky acoustic wave whose amplitude strongly depends on the propagation conditions of the guided wave. This, then, can reveal any delaminations and, potentially, regions having a weakened ground [5, 6].

OTHER NON-CONTACT ULTRASOUND METHODS

A variety of non-contact ultrasound techniques is currently available [5] including pulse laser generators [38, 39], optical interferometer detectors [40], and electromagnetic acoustic transducers (EMAT generators and detectors) [41-42]. Pulsed laser generators appear to be the most efficient as with a single pulse they can generate longitudinal waves, shear waves and surface Raleigh waves. The generated sound is usually detected by capacitive transducers in a non-contact fashion. The laser pulse may be easily delivered to remote or difficult locations to reach by means of an optical fibre. It should be noted, however, that in order to avoid damage to the object, the energy delivered to surface has to be limited. This is particularly true for artwork such as canvas and wood panel paintings; for sculptures and frescos this is not an issue. Similar to X-ray tomography, ultrasonic tomography can provide images of any desired layers through a solid object. However, the information that is obtained is distinctly different. This technique is widely applied to metals (in frequency ranges higher than 1 MHz). Also, a few tomographic investigations using low frequency transducers (20 kHz and 70 kHz) have been successfully performed on historical masonry structures. These are related to the Cathedral of Nicosia, located in a XV century small town in central Sicily. In this investigation it was shown that it was possible to detect the presence of damaged regions (plaster detachments, cracks and inhomogeneities) which correspond to the regions of the image where the signal has low velocity (these variations of the velocity depend primarily on the dynamic elastic moduli of materials).

PROTON-INDUCED X-RAY EMISSION

Proton-Induced X-ray Emission (PIXE) method where beam of protons is used to excite the atoms of an investigated object was recently used to identify pigments [4, 44]. The incident photon removes an electron from the inner orbits in the atom, leaving a vacancy on that orbit. The upper electron then relaxes to that vacancy, emitting a photon with energy equal to transition. This photon can be detected with a Si(Li) semiconductor detector. Since the electron configuration is specific for every element, the series of detected photon energies is specific to a particular element. Thus, PIXE can identify the various elements contained in pigments. The measured energy values then serve to identify the specific elements contained in the pigment, with the number of photons being proportional to the quantity of the element.

X-ray emission can also be stimulated by an X-ray itself, or by an electron beam. X-ray-stimulated emission is called *X-ray fluorescence* (XRF) and, unlike PIXE, it does not require a huge particle accelerator. In fact, an experimental XRF device requires only a source of X-rays and a detector. Owing to good penetration of X-ray through the painting materials, all pigments at all depths can be analyzed at the same time. Also, the entire testing procedure can be carried out in air [4]; this does, however, restrict the detection of elements from Na to S due to the absorption of X-ray radiation by these atoms by the ambient atmosphere [45]. Both PIXE and XRF can be successfully used to identify paint pigments [4, 45-47].

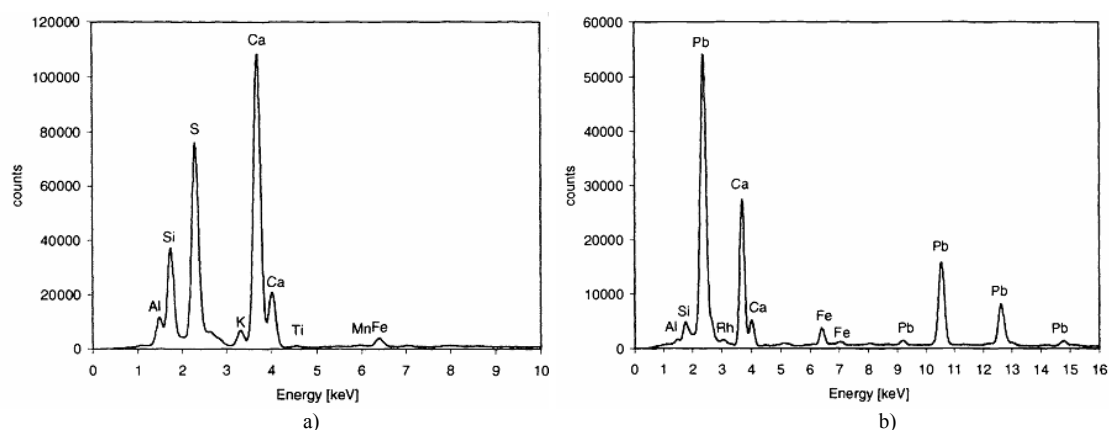


Fig.7 An example of PIXE spectra of a) pure ultramarine (Na, Al, Si, S, Fe) and b) ultramarine mixed with lead white [45].

For the study of a crystal lattice, X-ray diffraction (XRD) technique can be deployed. This method represents the diffraction of X-radiation by the periodic structure of solid media (which can be treated as a two-dimensional diffraction grating where the distance between atoms is comparable with the wavelength of X-radiation). In most of cases this technique is applied to a powdered sample, i.e., it requires the fragmentation of a painting or statue, making XRD a destructive method. On the other hand, it gives information on the crystal structure of the media, rather than on the presence of particular molecules or particular chemical elements. For this reason XRD is a good probe for testing the crystal structure of samples, which generates unique information with respect to the methods described so far in this article. In some cases XRD can help detect changes in the ground and/or paint structure with time, which is often associated with deterioration [7].

RAMAN SPECTROSCOPY

In 1928 two groups of scientists in India and USSR independently discovered a phenomenon, later named *Raman scattering* (in honor of one of discoverers). The main feature of this phenomenon is the appearance of additional frequencies in the optical spectrum of light

scattered by examined matter. It was shown that this resulted from the vibrational energy of the scattering molecules, giving additional frequencies to electrons, and in turn re-emitting the incident light. The vibrations are closely related to the intrinsic degrees of freedom of the molecule, or, in other words, they depend on its structure. This means that the scattering molecule adds its "signature" to the scattered light. Thus, a specialist who studies these spectra can recognize that signatures that are present and estimate which molecules are present in a studied sample. It is important to note that these "signatures" characterize the molecules rather than the atoms themselves (like PIXE or XRF, for example).

The method of Raman spectroscopy has found great use in the study of as paint pigments [8, 9], grounds [7], and varnishes. Specific spectral lines in the scattering spectra give information on the dyes used, whereas, the intensity of the lines corresponds to the proportions of the pigments. Using this information, an art specialist can reveal a fake or a later restoration. The retrieval of pigments with a well-known date of invention enables to date the artefact *post quem*. Other materials are known to have disappeared from the artists' palette, because they were substituted by others and retrieving enables to date artefacts *ante quem*. Finding anachronisms in the materials that were used, is a straightforward way for the exposure of counterfeit masterpieces. Another method consists of the comparison with the materials that were used in known works of the same artist. If it is well-established that a certain artist in a large group of works from a particular period never used certain pigments, finding these materials in a suspect painting deepens the suspicion and invites for further examinations. Additional lines in Raman spectra can also appear with time due to the deterioration of materials. This can be caused by changes in the chemical composition of material due to high concentrations of pollutants in atmosphere [7].



*Fig.8 Renishaw® Fiber optic Raman analyzer in operation.
University of Windsor, Materials and Surface Science Laboratory.*

The same method can be used for examining the deterioration of metal structures. For example, by examining the oxidization of bronze statues [48], one can extract information about the dominant chemical elements in the atmosphere surrounding the statue during its history, hence, revealing forgeries [49].

Our group is currently collecting Raman spectra for typical pigments used by painters all over the world. Naturally, each artist has his own preferences with respect to pigment selection;

this choice also varies from country to country as well. As such, the creation of this database will better enable us to determine information regarding the origin of a painting, the author and, even the locations where subsequent restorations took place [50-54].

CONCLUSION

The problems of art restoration, attribution, and forgery detection are increasingly important and may not easily be solved without employing modern scientific methods. There are numerous techniques that may be utilized for the examination of artwork. Unfortunately, only the following few are in active use by art researchers: X-ray, NIR visualizing, Raman spectroscopy and visual inspection. There is, however, great potential for using recently developed methods for scientific art analysis. Such methods as air-coupling technique, Lamb waves analysis, laser ultrasound, pulsed-phase thermography, and others can contribute significantly to art analysis science. Certainly they can be of great use in restoration, deterioration detection, and authentication of artwork, forgery detection, and attribution. The importance of incorporating advanced imaging algorithms for non-destructive artwork characterization and evaluation is evident. Let this paper serve as a challenge to researchers to continue developing better methods to evaluate art objects. These methods and techniques will enable researchers to collect critical information about the condition of artwork to make the art world more secure and transparent.

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