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Invited Talk at ISRP15

Radiation studies of items of cultural heritage

Heinz-Eberhard Mahnke[1,2,3]

[1]Ägyptisches Museum und Papyrussammlung, Staatliche Museen zu Berlin, Germany

[2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Germany

[3]Fachbereich Physik, Freie Universität Berlin, Germany

Corresponding author: mahnke@helmholtz-berlin.de

Abstract

When we see a piece of art or an archaeological object for the first time, a painting, a jewel, a sculpture or merely a simple piece of pottery on display in a museum or in an exhibition, we might be fascinated by its beauty or by its simplicity. However, many questions often arise in connection with such items on display, about its origin, its context, its provenance, its authenticity. Finding complete answers is a fully interdisciplinary task between art, humanities and natural sciences. With respect to the contribution from natural science, it often needs the application of just the best methods and techniques out of a multitude of possible options. Starting from a simple visual inspection with or without a magnifying glass the techniques reach up to highly sophisticated and advanced methods developed in atomic physics, nuclear physics and chemistry, such as available at large scale facilities, and even biophysics. In the following, a condensed review is presented on recent research on cultural heritage objects using various radiation techniques.

Introduction

For the understanding, restoration, and conservation of objects of our cultural heritage, the context from where they originate, their historical background, their intercultural relationship, their social context and many other aspects, it is absolutely needed to “look” underneath the surface of the object after a visual inspection to know all possible details about the object. Of course, it is mandatory, that such investigations are done non-destructively, or by minimal invasive means.

From visits to the museum, we all are aware of the sensitivity of precious paintings, manuscripts, or other delicate objects to the exposure of visible light, or of the part of the electromagnetic spectrum accompanying the visible light of our light source such as the UV or the infrared part of the spectrum. Similarly, we have to optimize the tools with which the objects are studied in terms of effectiveness in obtaining the information needed at minimal influence onto the object. The minimum requirement is that there should not remain any visible change on the surface.

A widely accepted tool in cultural-heritage investigations today is the well-known X-ray tube, which has turned into versatile handheld, easy to operate, portable instrument for X-ray fluorescence spectroscopy (pXRF) to determine *elemental composition*. Currently systems with 3-W power consumption, typically operating at 40 kV, are in use in field studies and museums magazines, some with focused beams of approximately 1-mm diameter so that even lateral distributions may be determined. For higher resolutions, well-collimated systems for the laboratory and for beamlines at synchrotron facilities are available, with the latter offering the advantage of monochromatic X-ray beams at adjustable energy. Due to the absorption length of the exciting X-rays and the absorption of the emitted fluorescence X-rays, information is limited to a typical thickness or depth of the order of around 100 µm of the material of the object, which of course is strongly energy dependent, less for lower energies and more for higher energies. If instead of x-rays charged particles are used to induce the emission of radiation, e.g. in the case of PIXE (**p**article **i**nduced **X**-ray **e**mission) at so-called "small accelerators" (like 2-MV tandems), the sensitive thickness is further reduced. If information from deep inside an object is wanted, then the use of neutrons could be a complementary alternative.

For *visualization* of inner structures of the artefact of interest, the exact determination of the elemental composition is not needed, provided the different parts of the structure yield enough contrast and the radiation is not totally absorbed by the object so that some fraction of the incoming intensity is transmitted and detected. In this review, the discussion is restricted to X-ray tomography. Other types like neutron tomography, magnetic resonance tomography (MRT) and others have already been applied to objects of cultural heritage, too. For *dating* purposes, radiation from inherently incorporated constituent atomic nuclei might be detected.

Common to all information obtained in natural science experiments, any experimental result needs a critical control of the conditions, premises and assumptions under which the result is obtained. Therefore, close cooperation between investigators and restorers is needed. And, wherever available, the background, the history of the artefact such as an excavation protocol or transfer protocol should be known.

Elemental Analysis

One of the simplest question centers around the problem of determining the elemental composition either as the major components or as trace elements. The solution for this problem is provided by characteristic relations well-known from atomic and nuclear physics. There is the emission of characteristic x-rays, after either the photo-electric effect, i.e. the absorption of x-rays, named X-ray fluorescence (XRF), or induced by particles, typically electrons, or protons or other light ions, named particle-induced X-ray emission PIXE. The x-ray energies roughly increase with the square of the elemental number Z (the so-called Moseley law (1)). However, XRF is not really possible for elements lighter than about sodium (Na) due to the so-called fluorescence yield, electron emission is the dominant emission mode for low Z elements. But for such elements, nuclear reactions are an applicable alternative: the so-called PIGE technique (proton-induced gamma ray emission). The various techniques are illustrated in Fig.1, where the nuclear techniques Rutherford backscattering spectroscopy (RBS) and the nuclear reaction analysis NRA with the emission and detection of a different particle are also sketched. From kinematics of the process, RBS can be used for near surface analysis with tens of nanometer resolution and with the surplus of simultaneous PIXE within the same experiment.

An early example for such elemental analysis in the case of trace elements, which also demonstrates the importance of the complementing interplay between (natural) science results and art historical information is the analysis of the ruby inlays in the Ishtar figurine from alabaster as described in the original publication by Calligaro et al. (2) and as reviewed within the context of archaeometry in refs.3 and 4. In the case of paintings and similarly in the case of palimpsests, the determination of a single element as representative for the used pigment may reveal hidden paintings (5) or erased and overwritten text (6), as discussed in the mentioned reviews, too. Fig.2 shows one of the very first revealing of an overpainted painting within van Gogh's "Patch of Grass". First demonstrated at the HASYLAB synchrotron facility at DESY, improvements in detection and mapping has lead to the development of a mobile system so that the equipment is now going to the museum and not the object to the large-scale facility.

Dating

In contrast to the use of induced emission of x-rays, as discussed above, the detection of radiation from built-in components of a cultural object can be used for dating or evidence for authenticity. In the case of the well-known C-14 dating method, the detection of the emitted radiation has been widely replaced by the direct counting of not-yet decayed C-14 isotopes in accelerator mass spectroscopy (AMS). With C-14 AMS, the sensitivity could be improved by up to six orders of magnitude reducing the needed amount for the sample accordingly. A famous example is the dating of the Theran eruption in the Aegean Sea (7). Within this publication, the difference between a so-called measured age and calibrated date is discussed, the correction applied reflects the variation of atmospheric concentration of C-14.

When the element lead Pb is part of a cultural object of interest, the alpha-decay of Po-210 following the decay of Pb-210 (half-life 22.3 years) may be used to distinguish manufacturing more or less as of present-day or many decades ago (8). This was applied to the fraud painting by Han van Meegeren (1889-1947) using lead white in paintings in the style of Jan Vermeer (1632-1675). In the case of the "Sky disk of Nebra", the absence of any alpha particles with the decay energy of Po-210 could be taken as an indicator for a genuine object, at least not manufactured during the last century (9). With C-14 dating on birchbark remains on associated finds, the disk could be dated to the bronze age.

Tomography

As stated in the Introduction, we restrict this review to X-ray tomography. While for the elemental analysis the succeeding emission of X-rays provides the fingerprint for studying objects of cultural heritage, the absorption itself via the photo-electric effect provides the means for imaging and reconstruction of the interior of an object. Here, revealing hidden text is our issue, as done in the case of papyri found on the island Elephantine near Aswan in Egypt. The title of the ERC funded project "Localizing 4000 Years of Cultural History. Texts and Scripts from Elephantine Island" indicates that we are dealing with several different epochs. Access to text is possible using absorption tomography when ink containing higher Z elements like iron Fe (Z=26), copper Cu (Z=28), or even lead Pb (Z=82) (e.g. ref. 10) provides significant contrast from the base material papyrus with carbon C (Z=6) as the major component of the organic material. The cross section for X-ray absorption increases with a high-power law with the element number Z as described in greater detail in refs. 11 and 12. The state-of-the-art is reviewed in ref.13, where similar projects like the Herculaneum project of carbonized papyri (due to the eruption of the Vesuvius) are referred to and references given as well. The setup used is a laboratory microCT system at the HZB, Institute of Applied Materials, as sketched in Fig.3. A summary of the recent unfolding of a package from the island Elephantine, now in the collection of the Musée du Louvre, written in Coptic is illustrated in Fig.4.

Many papyri, however, especially papyri written in earlier epochs before our era, are written with carbon ink, for which there is not sufficient contrast in the absorption-based tomography. For objects of this type, the development of suitable imaging techniques is imperative, a project we are currently working on.

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References

(1) H.G.J. Moseley: *The high-frequency spectra of the elements*, Phil. Mag. 26 (1913) 1024-1034

(2) T. Calligaro, A. Mossmann, J.-P. Poirot, G.Querré: *Provenance study of rubies from a Parthian statuette by PIXE analysis*, Nucl. Instr. Methods Phys. Res. B136-138 (1998) 846-850

(3) H.-E. Mahnke: *Nuclear physics methods in cultural heritage research - accelerators for art*, Acta Physica Polonica B45 (2014) 571-588

(4) H.-E. Mahnke, A. Denker, J. Salomon: *Accelerators and x-rays in cultural heritage investigations*, Comptes Rendus Physique 10 (2009) 660–675

(5) J. Dik, K. Janssens, G. van der Snickt, L. van der Loeff, K. Rickers, M. Cotte: *Visualization of a Lost Painting by Vincent van Gogh Using Synchrotron Radiation Based X-ray Fluorescence Elemental Mapping*, Anal.Chem. 80 (2008) 6436-6442

(6) U. Bergmann: *Archimedes brought to light*, Phys. World 20 (11) (2007) 39-43

(7) M.H. Wiener, J.W. Earle: *Radiocarbon dating of the Theran eruption*, Open Journal of Archaeometry 2014; volume 2:5265, pp.60-64

(8) B. Keisch: *Dating Works of Art through Their Natural Radioactivity: Improvements and Applications*, Science 160 (1968) 413-415

(9) E. Pernicka: *Archäometallurgische Untersuchungen am und zum Hortfund von Nebra*, in: Meller, Harald und Bertemes, François (Hrsg.), *Der Griff nach den Sternen. Wie Europas Eliten zu Macht und Reichtum kamen* (Tagungsbericht, Halle 2010) 719-734

(10) T. Arlt, H.-E. Mahnke, T. Siopi, E. Menei, C. Aibeo, R.-R. Pausewein, I. Reiche, I. Manke, V. Lepper: *Absorption edge sensitive radiography and tomography of Egyptian Papyri*, Journal of Cultural Heritage 39 (2019) 13-20

(11) D. Baum, N. Lindow, H.-Chr. Hege, V. Lepper, T. Siopi, F. Kutz, K. Mahlow, H.- E. Mahnke: *Revealing hidden text in rolled and folded papyri*, Appl. Phys. A (2017) 123:171

(12) H.-E. Mahnke, T. Arlt, D. Baum, H.-C. Hege, F. Herter, N. Lindow, I. Manke, T. Siopi, E. Menei, M. Etienne, V. Lepper: *Virtual unfolding of folded Papyri*, Journal of Cultural Heritage 41 (2020) 264-269

(13) H.-E. Mahnke, V. Lepper: *Virtual unfolding of folded Papyri*, Acta Physica Polonica B51 (2020) 541-549

(14) N. Kardjilov, A. Hilger, I. Manke: *CONRAD-2: Cold Neutron Tomography and Radiography at BER II (V7)*, Journal of large-scale research facilities A98 (2016) vol.2, http://dx.doi.org/10.17815/jlsrf-2-108

Figure Captions

Fig. 1:

Sketch of the ion induced processes for various analytical techniques. Labelled in red are processes based on nuclear interaction, while atomic processes are labelled in green.

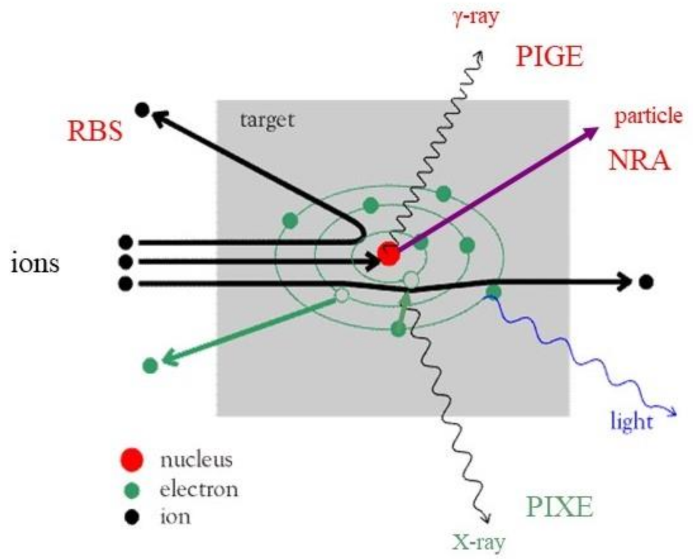

Fig. 2:

XRF mapping with monochromatic synchrotron radiation of the painting "Patch of Grass" by van Gogh. The insert shows the revealing of a portrait of a woman, already from the mapping of the antimony Sb X-ray intensity (see the red arrows). We thank the authors of that work to let us use their data (5) for this review.

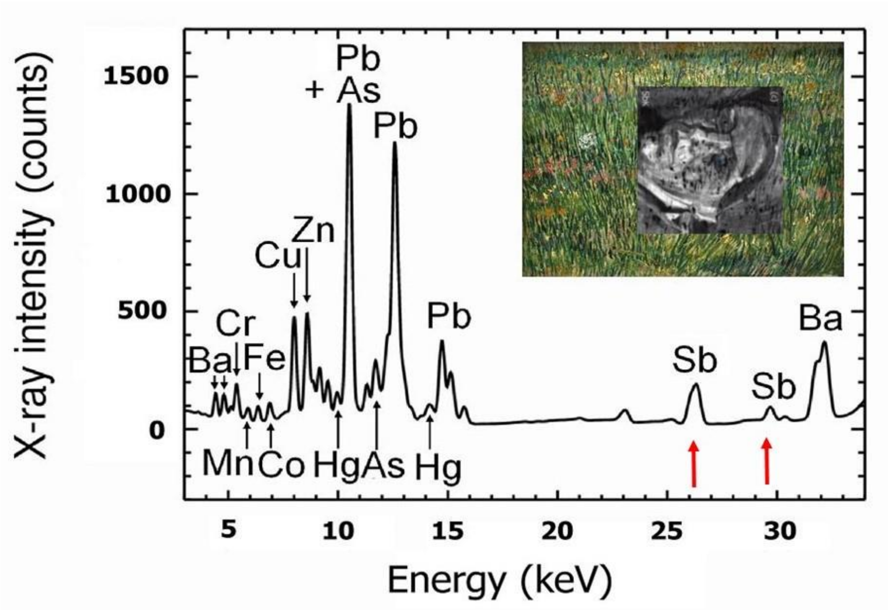

Fig. 3:

Setup of the µCT system. The voltage at the x-ray tube was adjusted for optimized absorption or transmission, resp. (courtesy of T. Arlt, A. Hilger, N. Kardjilov and I. Manke, see (14)).

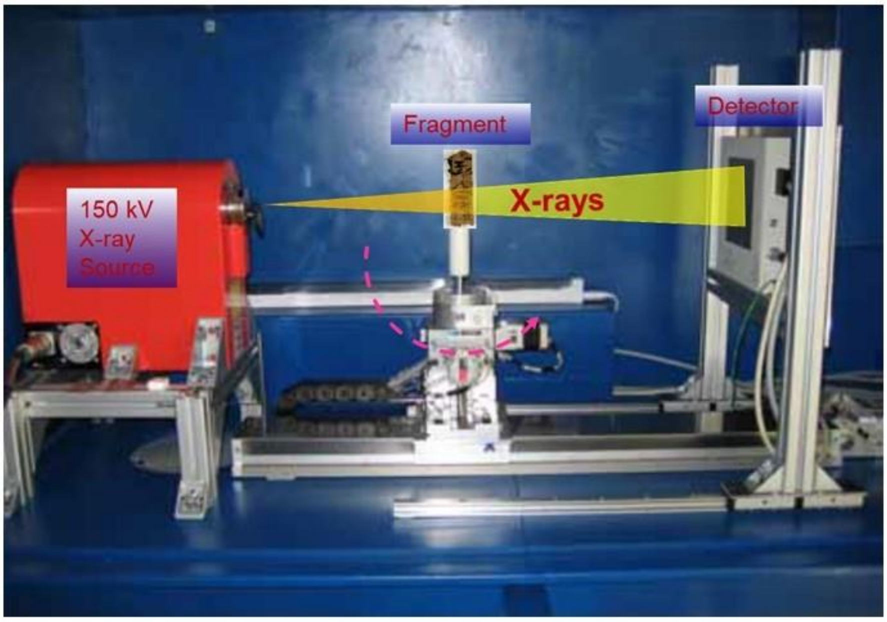

Fig. 4:

X-ray absorption tomography of a Coptic package L/El227b/1-pC. (left) Photo showing the "bandaged" package (with Japanese tissue), (center) volume rendered image, (upper right) cross section and marked central line for unfolding, (lower right) 2D-projection showing a Coptic word for (the) "Lord".