DISCOVERING NEW INFORMATION FROM HISTORICAL ARTEFACTS
USING ELECTROMAGNETIC RADIATION AND CHARGED PARTICLES AS A PROBE

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Archaeological artefacts included in cultural heritage contribute to the knowledge of our roots, which may help us to learn about our future. Although the connection between ancient times and nuclear technology seems farfetched, this paper will try to show how nuclear radiation of various kinds can be irreplaceable in the elemental composition analysis of an archaeological find.
The first steps and the present days

Scientific methods in archaeology and art began to be systematically applied in the late eighteenth century by the German scientist Martin Heinrich Klaproth (1743–1817), who published the first-ever quantitative analysis of an alloy of some Greek and Roman metal coins. Much later, in the early 1960s, various types of ion beam analyses (IBA) were developed and put into routine use. Further developments of IBA-based analytical methods were related to progresses in low-energy accelerators, in detectors for particle, X-ray and γ-ray and in systems for processing experimental data [1].

Ion beams of several MeV, produced by small accelerators, penetrate into matter, interact with the atoms of the sample and produce, among other phenomena, X-rays and γ-rays, which carry information about the investigated artefact. The small accelerators can provide a wide range of ion beams (protons, alphas and heavy ions), with flexible energy range (and thus adjustable probed depth) and diameter of the beam (from millimetre to micron size). They can thus provide tailored tools for the study of the diverse objects of Cultural Heritage.

Nuclear analytical methods successfully applied in archaeometry

Archaeometry involves non-invasive surveys of the terrain, science-based dating methods and analytical techniques for object characterization. Nuclear physics contributes significantly to the dating methods (radiocarbon dating) and to analytical methods with techniques sensitive to practically all the elements of the periodic table and capable of reconstructing the spatial distribution of the elements present in the sample [2, 3].

It should be emphasized that applications of nuclear analytical methods on the cultural heritage have recently been collected for a new, topical paper published by the Nuclear Physics Board of the European Physical Society, of a few highlights of which we would like to present here.

How ion beam methods work

Rutherford backscattering spectrometry (RBS) is an analytical non-destructive method which is based on the measurement of the energy spectra of MeV ions (protons, He⁺, Li⁺, or heavier ions) elastically scattered from solid samples [2, 3]. From the analysis of the backscattered particles’ energy distribution it is possible to determine the elemental amounts in the sample and their depth distributions.

When ions and matter interact via nuclear reactions, charged particles and/or γ-rays are produced (see Figure 1). The Nuclear Reaction Analysis (NRA) technique is based on the study of the energy spectrum of these charged particles and γ-rays. The yield of nuclear reaction products is proportional to the reaction cross sections (which define the probability of each type of interaction) and the density of target atoms in the sample. Most frequently used reactions are (p,α), (d,p), and (d,α), which allow indicating the presence and the concentration of several isotopes typically from H to 32S, such as 13C, 14C, and 16O [2-4], just to name a few. Energy loss by the incident ion can be used to determine depth profiles by resonance scanning using a (p, γ) reaction, where γ-rays are detected. Particle-Induced Gamma Emission (PIGE) is based on nuclear reactions, most typically (p, γ), induced in specific isotopes. The energy of the γ-ray lines indicates the elements, while the intensity is related to their concentrations.

Particle-Induced X-ray emission (PIXE) exploits X-ray emission for elemental analysis [2, 3]. The energy of a peak in the X-ray spectrum is specific for a particular element, and its intensity is proportional to the elemental concentration. PIXE has a very low detection limit, down to several ppm in standard practice.

A big technological progress was made after the ion microbeam development. In a microbeam, the ion beam from the accelerator passes through a lens (a combination of magnetic quadrupoles with alternating polarities) focusing the high energy ions. The samples are irradiated with an ion beam focused onto a spot that can be as small as a few hundreds of nm in diameter. Standard IBA techniques are used to characterize the irradiated object. By raster-scanning the beam over the sample surface, a 2D or 3D distribution of elements can be determined with nm depth resolution and lateral resolution limited by the size of the beam spot, see Figure 2 [4, 5, 6].

Some archaeological artefacts cannot be placed in a vacuum chamber because of their large size or the presence of volatile components. Such samples can be analysed using an external ion beam. The beam is extracted from the evacuated beam line into air through a thin window, made of thin metal foils, strong plastic materials like kapton, or Si₃N₄. Practically all setups now allow the scanning measurement mode that produces elemental
concentration maps. The target is surrounded by an array of detectors. Normally there are at least two X-ray detectors, one with a thin window detector for soft X-rays and a detector with a large solid angle (but equipped with an additional absorber) for hard X-rays. The external microbeam set-up can be improved to be versatile and allows all IBA techniques to be used individually or in combination [6], namely PIXE–PIGE–RBS with protons, PIXE–PIGE–NRA with deuterons, PIXE–RBS with He⁺ ions [1-6].

**Examples of the archaeological artefacts analyses**

The example of metal analysis demonstrates the identification of the gilding technique, see Figure 3. The object studied comes from the Early Medieval Age, which favoured gilded silver or bronze jewellery. The methods applied were differential PIXE and RBS with in-air proton beam [6]. Differential PIXE is based on the sequential measurement in the same spot such that protons reach different target depths. This is achieved by the variation of the proton incident angle or by the variation of proton energy. The results of the de-convolution procedure are concentration profiles, which can reach up to a few tens of microns below the target surface. Figure 3 shows the elemental concentration profiles of a gilded strap end [7, 8].

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The amount of additional elements (Cu, Hg) in the gold layer undoubtedly reveal the amalgamation gilding technique used.

Examples of elemental composition study of precious stones

PIGE was very efficiently used for light elements and PIXE for medium and heavy elements in qualitative and quantitative analysis of emeralds and garnets, popular among Romans and their barbaric successors. Emeralds contain a known fraction of beryllium, which can be measured either directly or taken into account numerically for the calculation of matrix effects. The provenance sites of the precious stones are certainly of interest as they indicate the extent of trade routes established by the Romans [9, 10] and can be determined via various trace element concentrations in the precious stone. The fluid channels in a set of emeralds excavated in a Roman grave from Slovenia point to a source in Egypt, while emeralds from another grave may be traced to Afghanistan.

Another interesting application is the study of the origin of Lapis lazuli. Lapis lazuli is a semi-precious blue stone widely used for different purposes since the antiquity. However, at present there are still some lacking pieces of information about its trade in ancient times [11, 12, 13]. An external proton microprobe was used, as the external beam allows for non-invasive, multitechnique (PIXE, PIGE and ionoluminescence IL) study of objects of almost any shape and dimension, see Figure 4. For the provenance discrimination the study focused on markers, such as for example the presence or absence of the trace elements in the stone of a specific mineral phase. After this study, some of the markers found on rocks have been successfully used to identify the origin of six precious artworks.

Conclusions

The application of atomic and nuclear techniques in the study of archaeological objects provides a historian or archaeologist with ‘material’ information that can help in the understanding of the way of life in ancient times. This knowledge is necessary for the testing of the authenticity and provenance of artefacts and for the preparation and implementation of the necessary restoration work. All of these objectives are common to the very large community of people working in the field of archaeometry, i.e. the ‘application of science to art and archaeology’.

About the Authors

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Lorenzo Giuntini is an applied nuclear physicist, one of the founders of the LABEC, the Florence laboratory of the National Institute of Nuclear Physics for the study of cultural heritage and environment by nuclear techniques. He has been the initiator of the Florence microbeam and his main expertise are external microprobe, IBA and XRF. He presently serves as one of the two responsible of the Tandem laboratory in Florence and is associate professor of experimental physics at the University of Florence, Italy.
References


Letter to the Editors

by Wolfgang Kundt,
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Dear Editor,

Today I approach you because of the science-fiction contribution on GW140915 by Angela Di Virgilio, in your issue 47/2 (2016) on pages 9–10, claiming that this GW event was launched by two stellar-mass black holes. It ignores a number of serious results during the past 7 years, by Pankaj Joshi, Bahram Mashhoon, Hernando Quevedo, Daniele Malafarina, and even Stephen Hawking (on 24 Jan. 2014), having proved that BHs are no longer expected, as results of gravitational collapse. It was derived by applying the chirp mass formula from John Wheeler’s published lecture notes to the emission event, an analytical formula which has been derived for 2 point masses circulating around each other at large separations, and successfully applied to several binary neutron stars, but whose validity expires as soon as two stars approach each other close enough for tidal deformations, and for the emission of strong gravitational waves. The emitting masses then turn ill-defined. Instead, above gravitational signal has most likely been emitted by two coalescing neutron stars, at a (smaller) distance of <~ 30 Mpc, a fate which some day likewise be shared by the Nobel (cf. https://wolfgangkundt.wordpress.com).

The author responds

Dear Prof. Kundt,

Some more exotic interpretations of the GW140915 event are possible, but the interpretation given by the LIGO/Virgo collaboration is probably the most economic and simple at the current stage of our knowledge. I’m more in the experimental side and my personal reaction in front of this event is that the antennas must be improved in the low-frequency part of the spectrum. I’m not the right person to fully address and reply your question; the GW community is rather large and this question need to be addressed to the right people. In any case, in order to give you a quick answer, I have discussed a bit with my colleagues and in the following I report some comments.

The claim that BHs have been proved not to exist is a bit misleading in this context. For example, the Hawking opinion you cited is mainly concerned with the real, absolute “blackness” of BHs, which is quite unrelated with the behaviour of these maybe “not completely black” objects when they collide. All these alternatives, naked singularities and so on, are surely interesting and worth to be experimentally tested.

Coming back to GW140915, trying to avoid too speculative physics, or as you said science fiction, for sure there is a clash between the observed signal (its frequency and its duration) and your suggested interpretation as a pair of neutron stars. For a NS-NS coalescence the signal is expected to remain in the LIGO sensitivity band much longer, its duration) and your suggested interpretation as a pair of neutron stars. For a NS-NS coalescence the signal is expected to remain in the LIGO sensitivity band much longer, mainly because a NS is much more compact that a 30 solar mass BH. This prediction is not based on the quadrupole approximation you are referring to, but on the analytical post Newtonian expansion and, in the final stage, on numerical simulation of full General Relativity. Quite remarkably, there is an overlap region where the two approaches agrees very well.

In any case, my personal hope is to have soon GW antennas on and with improved sensitivity, especially at low frequency, in order to help clarifying all this complicated matter.