

**NEUTRON RESONANCE IMAGING INSTRUMENTATION AT THE ISIS  
NEUTRON SOURCE**

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**ABSTRACT**

A modular system for neutron resonance capture imaging (NRCI) and neutron resonance transmission (NRT) has been designed and constructed for the ANCIENT CHARM project. The main components of the NRCI/NRT facility are: (1) a pencil beam collimation; (2) a gamma detector bank; (3) a neutron transmission detector; and (4) moving sample system. Here we report on the installation of the components on the Italian Neutron Experimental Station (INES) at ISIS. An assessment of the system performances on the basis of the preliminary results is presented.

**1. Introduction**

ANCIENT CHARM [1] is a EU-funded project which aims to develop and apply novel non-destructive neutron-based material analysis techniques. The main applications are envisaged for cultural heritage (CH), but isotope-resolved images can be used for a variety of other applications, like for instance estimation of homogeneity of nuclear samples. ANCIENT CHARM aims to combine the information provided by traditional non-destructive methods based on thermal neutrons, like prompt gamma activation analysis (PGAA) [2] and neutron diffraction (ND) [3], with relatively new methods based on neutron resonance absorption in the epithermal region. New instrumentation aimed to perform element- and isotope-sensitive neutron radiography in the epithermal energy region has been tested at the ISIS and GELINA pulsed neutron sources. This instrumentation makes use of the time-of-flight (TOF) technique available at pulsed neutron sources to measure the resonances occurring in the neutron capture cross sections. Absorption resonances are characteristics of different isotopes, and can thus be used as a fingerprint for elemental identification. ANCIENT CHARM instrumentation conjugates, in a modular set-up, two approaches for identifying elements into a sample through neutron resonances analysis, into an imaging technique called *neutron resonance capture imaging combined with neutron resonance transmission* (NRCI/NRT). With the newly developed equipment it is possible to obtain information that is both element-resolved and space-resolved, i.e. allows mapping of the element distributions inside a sample.

## 2. Principles of NRCA/NRT

Neutron absorption resonances occur for a large quantity of isotopes in the epithermal energy range ( $E_n$  up to 1 keV). Direct spectroscopy with neutrons in this energy range is difficult; as a consequence, the TOF technique available at pulsed neutron sources is better suited for the detection and identification of resonances. Resonances appear as characteristic dips in the total neutron cross-section. Energy position of the resonances can be identified via the well known relation:

$$E_n = \frac{1}{2} m_n \left( \frac{L}{t} \right)^2 \quad (1)$$

where  $m_n = 1.675 \times 10^{-27}$  kg is the neutron rest mass,  $L$  is the neutron flight path from the source to the sample and  $t$  the time to cover it. Following neutron capture, the nuclei are usually left in an excited state and they de-excite through the emission of a prompt  $\gamma$ -ray cascade. These  $\gamma$ -rays can be detected with suitable detectors, and the time stamp of the photon emission is thus recorded, allowing for measuring the neutron energy and thus identifying the absorbing isotope. This is the basis of the neutron resonance capture analysis (NRCA) technique [4], that has given interesting results in the field of cultural heritage [5], being especially sensitive to some elements of archaeological interest (for instance Cu, Ag, Sn, Zn, As, Au).

Standard NRCA, is *a priori* non position-sensitive; in order to obtain images it is necessary to scan the whole sample with a collimated neutron beam. Neutron resonance transmission technique (NRT) on the other hand, utilizes a 2D position-sensitive neutron detector (PSND) in transmission to collect two-dimensional maps of resonance data thus making use of the whole incident neutron beam. Figure 1 illustrates the operation principle of NRCA and NRT. For NRT the neutron beam transmitted through the sample is recorded

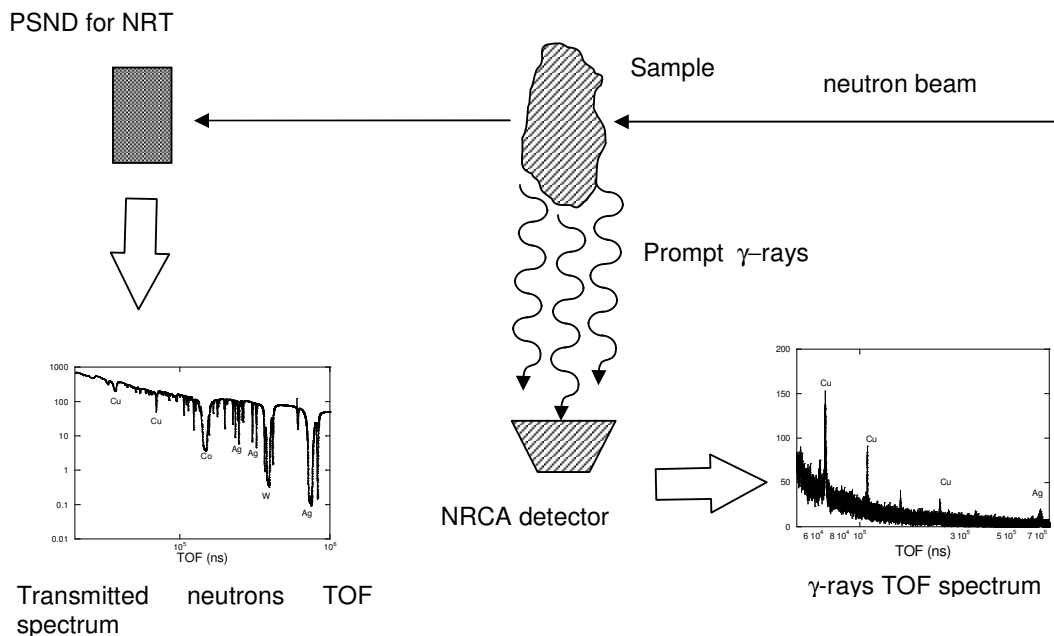


Figure 1: operation principles of NRCA and NRT.

as a TOF spectrum; the resonances appear as characteristic dips. Energy position of the resonances are identified by they time-of-flight. While being less sensitive than NRCA for some elements, NRT gives the advantage of directly obtaining 2D space-resolved information in a shorter time.

ANCIENT CHARM instrumentation combines a radiography-type set-up for NRT with a  $\gamma$  detector bank and neutron collimation system, the latter intended to allow precise pinpointing of selected regions of the sample to be analysed with NRCA.

### 3. The ANCIENT CHARM set-up

#### 3.1 The NRCA/NRCI set-up

Figure 2 illustrates the main components of the NRCI/NRT device as installed at the INES beamline [6] of ISIS. The INES beamline was chosen because of its relatively long flight path (about 23 m), allowing for good TOF resolution, and its flexible layout. Although the INES neutron beam is thermalised, it still provides a reasonable flux of epithermal neutrons (up to  $10^6$  neutrons/scm<sup>2</sup> at 100 eV neutron energy, providing a 180  $\mu$ A proton current is provided). Because neutrons in the interesting energy region for NRCI and NRT are little influenced by air, the INES vacuum tank is not evacuated for measurements; analysis in air is also beneficial for those fragile CH samples (for instance archaeological findings) which are not in danger of being damaged by sitting in a vacuum.

The  $\gamma$ -ray detector bank (figure 3) contains 28 Yttrium-Aluminium-Perowskite (YAP) scintillator detectors, coupled to photomultiplier tubes (PMTs) and stacked on an ideal cylindrical surface surrounding a column of 200 mm diameter reserved for the sample movements. The detectors are mounted in three support rings made of lithium carbonate ( $\text{Li}_2\text{CO}_3$ ) in an epoxy resin matrix. As it is known,  ${}^6\text{Li}$  has a relatively high neutron absorption cross section via a reaction that does not produce  $\gamma$ -rays. Thus,  ${}^6\text{Li}$ -rich materials are good candidates for a structure that combines both support and neutron shielding. The bottom and top ring each carry a set of 10 detectors; the mid-ring (which has cut-outs to allow for the neutron beam to pass through supports) 8 detectors.

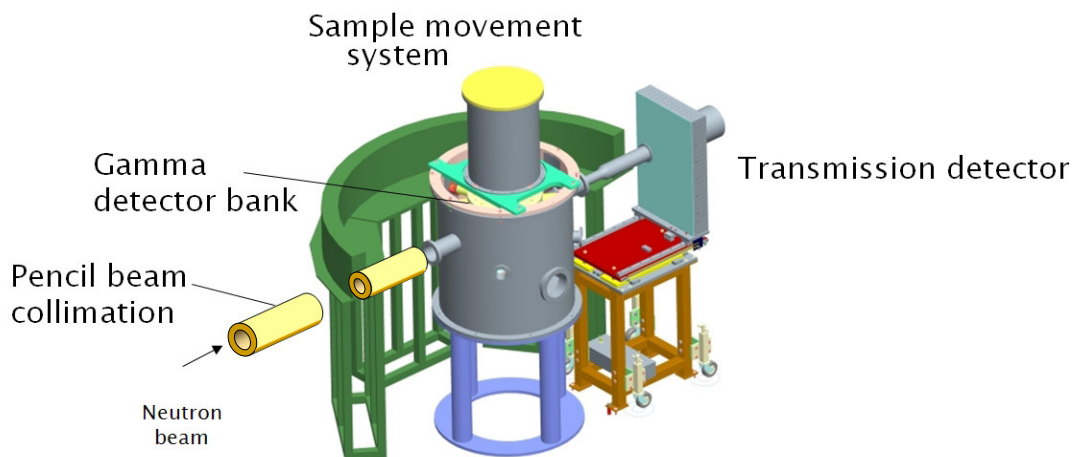


Figure 2: drawing of the main ANCIENT CHARM components installed inside the INES blockhouse. The crescent-shape green structure is the diffraction detector bank for normal operations of INES as a diffractometer.

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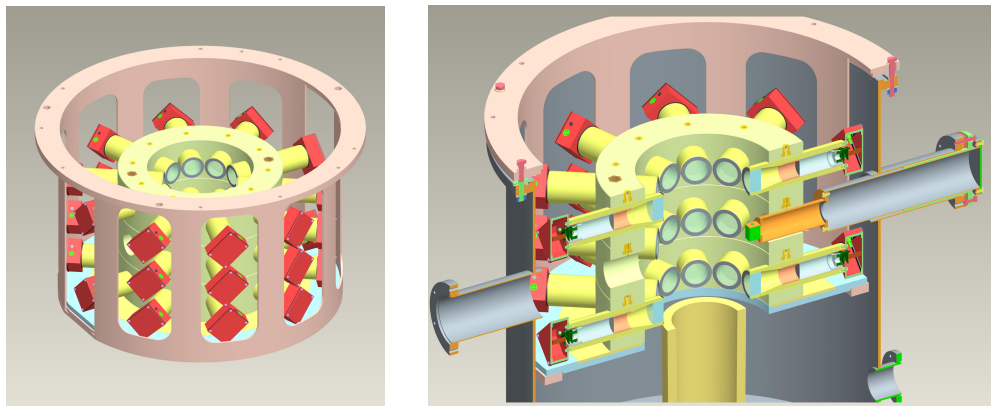


Figure 3: drawings of the  $\gamma$  detector bank. Left: the bank with the 28 detectors and preamplifiers (red) into the aluminium “basket” support. Right: section of the detector bank inserted into the INES sample tank. The cylindrical space left for sample movement is 20 cm diameter.

The choice of the best  $\gamma$ -ray detectors has been the subject of tests done both at ISIS and at GELINA [7] in order to compare different detector systems and test the feasibility of NRCI in realistic conditions. YAP detectors were found as the most promising, mostly due to i) absence of intense neutron capture resonances in the interesting energy range, ii) fast time response (fast scintillation decay constant  $\tau = 27$  ns), iii) relatively high light output (about 18 photons/keV), iv) acceptable detection efficiency in the required energy range.

The necessity of stacking together many detectors in a limited space can be the origin of cross-talk problems.  $\gamma$ -rays emitted after neutron capture in the sample can be scattered by a solid component of the bank into a detector and then recorded. If the solid component that produces the scattering is another detector, it can result in the same photon to be counted twice. This phenomenon is known as *cross-talk* and can possibly be the cause of distortion of the recorded signal. GEANT4 [8] simulations have been performed in order to estimate the amount of this effect in the NRCI detector bank. Results have shown that cross-talk can be kept at a negligible level, especially with the use of a suitable low level discrimination threshold [7].

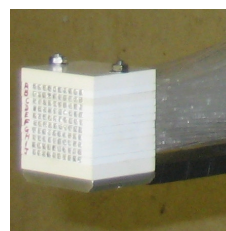
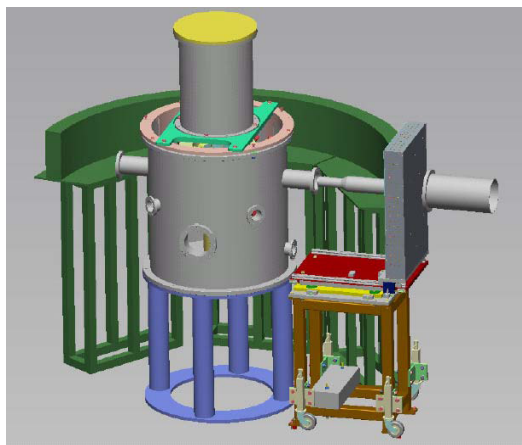


Figure 4: left: drawing of the PSND setup inside the INES blockhouse. Right: picture of the ANCIENT CHARM PSND

The NRCI collimator consists of two sections, each made  $\text{Li}_2\text{CO}_3$  in resin (Figure 2). Collimator Section 1 consists of 8 segments with central bore holes of varying diameters, designed following the classical scheme of the *irises* [9]. A Pb element is available to optionally replace one of the lithium resin elements. Section 2 is made of 4 segments. Of the two sections in which the collimator is made, Section 1 is placed inside the INES shutter, while Section 2 is installed in the blockhouse, approximately 1.5 m upstream from the INES sample position. The orientation and positioning of Section 2 can be fine-tuned using an xy-motorised table and alignment screws. The combined collimator sections are designed to produce a 5 mm beam spot of epithermal neutrons at the sample position.

### 3.2 The NRT set-up

A 100-pixel PSND is installed in transmission geometry at the beam exit of the INES tank (Figure 4). Every pixel is a GS20 Li-glass crystals ( $2 \times 2 \times 9 \text{ mm}^3$ ), mounted on a pitch of 2.5 mm, resulting in a  $25 \times 25 \text{ mm}^2$  active area. The Li-glass pixels are embedded in a support frame made of boron nitride; this material was chosen because of its neutron-absorbing properties and mechanical characteristics. Each pixel is coupled, via a 5 mm thick glass disperser and a bundle of four 1 mm diameter acrylic fibre optics, to a 16-channel PMT. 7 PMTs are required to fibre-couple a total of 100 pixels. The PSND was based on previous experiences with a cold-neutron PSND [10] and on a 16-pixels prototype PSND optimised for the epithermal energy range [11]. The novelty of the ANCIENT CHARM PSND is the increased neutron efficiency (about 25% at 10 eV and 4% at 1000 eV) due to higher pixel depth, allowing to measure a broad neutron energy range. For high neutron energies the scattering effects in the material surrounding the pixels become more relevant than for a cold neutron detector. The design of the new PSND has been supported and validated by GEANT4 simulations in order to assess and minimize the effect of neutron scattering on the cross talk behaviour for different combinations of support, neutron absorbers and fiber optics materials [12].

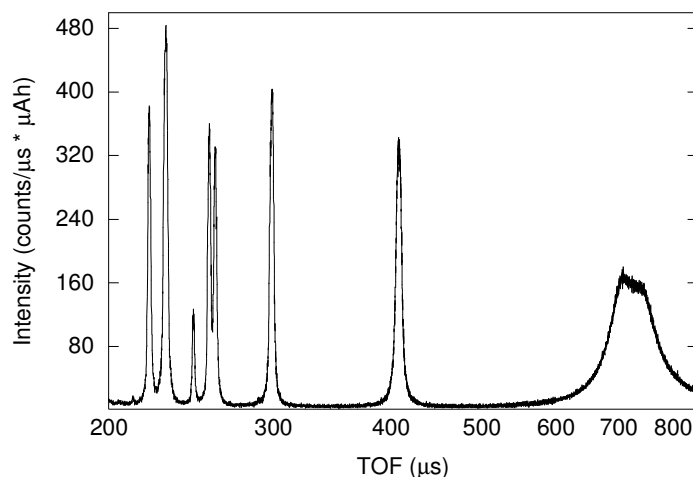


Figure 5: Example of a NRCA spectrum from a 0.2 mm silver sample acquired for 300 s. All the resonances visible in the spectrum are due to silver isotopes. The TOF region in the plot represents the energy region between 4 eV and 200 eV.

#### 4. Assessment of imaging performances

NRCA performances of the  $\gamma$  detector bank was tested with measurements on metal samples (both pure elements and certified alloys). Preliminary results (Figure 5) show that elements with intense resonances below 1 keV can be observed immediately (measurement times of few minutes). Energy resolution is due to time resolution of the whole system. Intrinsic time resolution of the ISIS standard data acquisition electronics (DAE) is 62.5 ns. At energies above 1 keV, the resolution is dominated by the width of the (double) neutron pulse (about 300 ns). At lower energies the observed profile is mostly dominated by the natural width of the resonances [7].

The spatial resolution achievable with NRCI was tested combining the detector bank with the collimator and positioning device to scan a reference sample ( $2 \times 2$  mm<sup>2</sup> In or bronze) and determining the intensity of selected resonances as a function of the scan variable. The measured FWHM of the almost circular beam spot is about 3.3 mm at 9 eV (<sup>115</sup>In) and 3.9 mm at 230 eV (<sup>65</sup>Cu) (Figure 6).

For what NRT is concerned, the prototype detector, tested both at GELINA and ISIS, had shown promising results both in terms of efficiency and accuracy [11]. Traces of Ag could be quantified down to a concentration of  $10^{-4}$ . Moreover, the whole system showed a count rate capability of about 200 kHz/pixel with a 5% dead time. For pixelated detectors, the pixel pitch gives an absolute limit of space resolution, however other effects, like for instance the neutron beam divergence, can worsen effective resolution. Scanning of an In wire in 0.2 mm steps and recording the intensity of the 9 eV resonance showed an effective resolution of about 2.5 mm FWHM (Figure 7), thus showing that the system reaches the design performances.

#### 5. Conclusions

Aim of ANCIENT CHARM project is to conjugate “...*Neutron resonant Capture Imaging and other Emerging Neutron Techniques...*” for non-destructive analysis of materials with main applications in cultural heritage, where non-destructiveness is often a

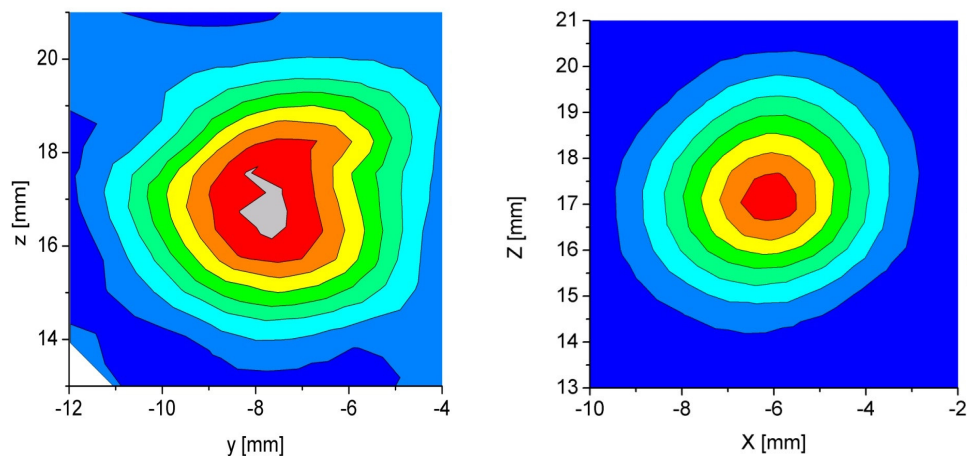


Figure 6: Collimated epithermal neutron spot at INES sample position. Both images have been recorded by scanning a  $2 \times 2$  mm<sup>2</sup> sample of bronze and indium, respectively. Left: image of the 230 eV resonance from <sup>65</sup>Cu. Right: image of the 9 eV resonance from <sup>115</sup>In.

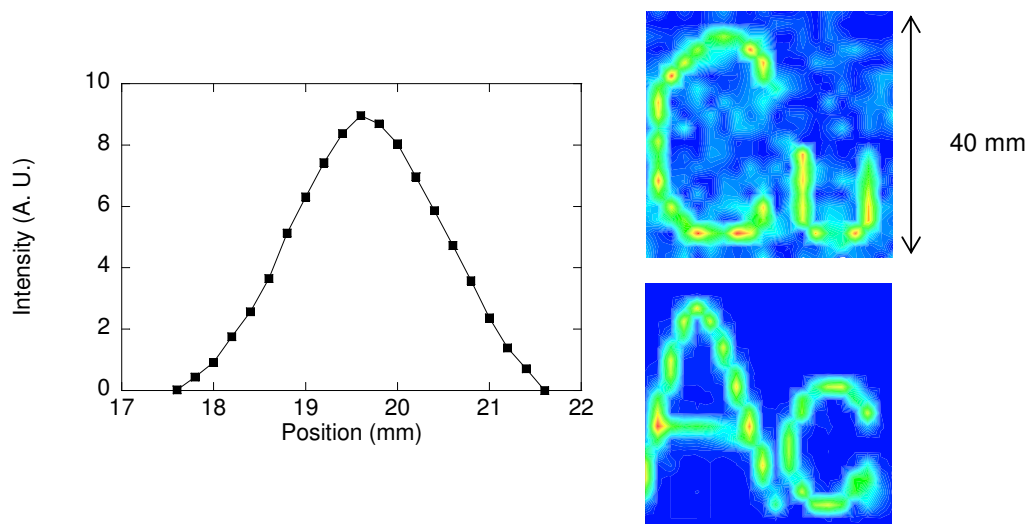


Figure 7 Imaging performance of the NRT system. Left: intensity of the 9 eV resonance recorded in a single pixel in a 0.2 mm step scan of a pure indium wire in front of the PSND. The FWHM is about 2.5 mm. Right: transmission images of the 230 eV resonance from copper (top) and 5 eV resonance from silver (bottom). The metal wires used for producing the images are 1 mm diameter.

fundamental requirement. In this paper we have described the NRCI/NRT equipment developed during the project and available for experimenters at the INES beamline at ISIS. The development of the analysis procedures is still in progress, but the first tests and experiments have given encouraging results, especially in terms of imaging capabilities. Both NRCI and NRT offer element-sensitive imaging capability, that is relatively fast, non-destructive and flexible. Moreover, with the use of a suitable scanning/positioning system, it can be developed into a 3D technique with standard tomographic procedures. The transmission and capture measurements give a joint indication of the sample content; the higher efficiency of the  $\gamma$ -ray detector bank allows for the determination of trace elements in the sample, while the PSND offers a rapid 2D mapping of the major components in the sample.

## 6. Acknowledgements

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## 7. References

1. G. Gorini, *Il Nuovo Cimento C* **30** No.1 (2006) 47.
2. G. L. Molnar (editor), *Handbook of Prompt Gamma Activation Analysis with neutron beams* (Kluwer Academic Publisher, Dordrecht, 2004).
3. W. Kockelmann, S. Siano, L. Bartoli, D. Visser, P. Hallebeek, R. Traum, R. Linke, M. Schreiner, A. Kirfel, *Appl. Phys. A* **83** (2006) 175.

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Grindelwald, Switzerland

4. H. Postma, M. Blaauw, P. Bode, P. Mutti, F. Corvi, P. Siegler, *J. Radioanal. Nucl. Chem.* **248** No. 1 (2001) 115
5. H. Postma, P. Schillebeeckx and R. B. Halbertsma, *Archaeometry* **46** No. 4 (2004) 635
6. S. Imberti, W. Kockelmann, M. Celli, F. Grazzi, M. Zoppi, A. Botti, A. Sodo, M. Leo Imperiale, M. de Vries-Melein, D. Visser and H. Postma, *Meas. Sci. Technol.* **19** (2008) 034003
7. E. Perelli Cippo, A. Borella, G. Gorini, W. Kockelmann, A. Pietropaolo, H. Postma, N. J. Rhodes, P. Schillebeeckx, E. M. Schooneveld, M. Tardocchi, R. Wynants, *A detector system for Neutron Resonance Capture Imaging*, accepted for publication in *Nucl. Instr. Meth. A* (2010)
8. S. Agostinelli *et al.*, *Nucl. Instrum. Meth. A* **506** (2003) 250
9. C. G. Windsor, *Nucl. Instrum. Meth. A* **243** (1986) 470
10. A. Steuwer, P. Withers, J. R. Santisteban and L. Edwards, *J. Appl. Phys.* **97** (2005) 074903-1
11. E. M. Schooneveld, M. Tardocchi, G. Gorini, W. Kockelmann, T. Nakamura, E. Perelli Cippo, H. Postma, N. Rhodes, P. Schillebeeckx and the ANCIENT CHARM Collaboration, *J. Phys. D: Appl. Phys.* **42** (2009) 152003
12. E. Perelli Cippo, G. Gorini, M. Tardocchi, R. Cattaneo, N. J. Rhodes, E. M. Schooneveld, T. Nakamura, P. Radaelli, W. A. Kockelmann, A. Pietropaolo and the ANCIENT CHARM Collaboration *Meas. Sci. Technol.* **19** (2008) 034027