Neutron Detectors Based Upon Artificial Single Crystal Diamond

Maurizio Angelone, Giulio Aielli, Salvatore Almaviva, Roberto Cardarelli, Daniele Lattanzi, Marco Marinelli, Enrico Milani, Giuseppe Prestopino, Mario Pillon, Rinaldo Santonico, Aldo Tucciarone, Claudio Verona, and Gianluca Verona-Rinati

***Abstract*—This paper reports about state-of-the-art artificial Single Crystal Diamond (SCD) neutron detectors based on a multilayered structure and grown by chemical vapour deposition (CVD) technique. Multilayered SCD detectors covered with a thin layer of ^{6}LiF allow the simultaneous detection of both slow and fast neutrons and can operate in pulse and current mode. These detectors can also be produced with a thin layer of Boron. Application of SCD detectors to neutron detection around fusion tokamak is reported. Some problems related to the processing of the very fast electrical pulse produced by diamond are addressed and the achieved and foreseen development of the processing electronics is reported as well.**

***Index Terms*—CVD mono-crystalline diamond, fast electronics, multilayered detector, neutron detector, sandwich detector.**

I. Introduction

DIAMOND is a very interesting material for radiation detection because it exhibits many outstanding properties such as: high band-gap (5.3 eV), high break-down field, high carrier mobility and high radiation hardness [1], [2]. These properties make in principle possible the realization of fast, low noise and radiation hard detectors [3], [4]. On the other hand, the recent development of the fabrication process of single crystal Chemical Vapor Deposition diamond detectors (SCD) in a p-type/intrinsic/metal layered configuration has allowed the production of devices with highly reproducible characteristics [5]. Under α-particle irradiation these SCD detectors have shown 100% charge collection efficiency and a resolution $<1\%$. Thanks to their excellent properties these SCD detectors were already tested in many different areas which range from UV to X-ray and from charged particle to neutron detection [6]–[8].

Since a very promising application of diamond is neutron detection a further step was the deposition of a thin layer of LiF enriched in ^{6}Li [5] on top of the detector. Such detector allows for the simultaneous detection of slow and fast neutrons and was successfully in use for almost two years at JET tokamak [9]. More recently the ^{6}LiF layer was replaced by a boron one and the response of such new detector is outlined in this paper together with the properties of a new type of detector developed for dosimetry and made by a "sandwich" structure (two layers of diamond with in between a ^{6}LiF or B layer). The boron covered detector is used for the Boron neutron capture therapy. The above detectors can operate both in pulse and current mode.

Under harsh working conditions, such as those expected in a fusion tokamak, an ideal working scheme is based upon locating the processing electronics far away from the detector. To transport far away the small and very fast signals produced by the diamond detector without loosing information is not an easy task to deal with. Special cables and a new and very fast electronics are thus necessary to transport and analyze the pulses produced by diamond detectors. This is under development in our laboratory and under testing at JET tokamak. Preliminary results are reported in this paper.

II. Diamond Detectors

A. Diamond Detector Production

The devices are fabricated at Rome "Tor Vergata" University by microwave chemical vapor deposition (MWCVD). Low cost $4 \times 4\ \mathrm{mm}^2$ commercial high pressure high temperature (HPHT) single crystal substrate is firstly covered by a highly conductive $(5\ \mathrm{Ohm} * \mathrm{cm})$ Boron-doped $(0.5 - 1. * 10^{20}\ \mathrm{at/cm}^3)$ CVD diamond film, whose thickness is usually about 10–20 μm. This layer is used as a back, grounded contact. An intrinsic diamond layer (thickness up to 200 μm), which constitutes the sensitive element to the ionizing radiation, is then grown on the doped surface in a separate clean reactor in order to avoid contamination of the intrinsic layer. On the top of the diamond a 2.5 mm diameter aluminium (or chromium) contact $(10^2\ \mathrm{nm\ thick})$ is then thermally evaporated. This multilayer geometry (Fig. 1) allows to separate the response of the high quality intrinsic CVD sensing layer from the one arising from the HPHT substrate, if

Manuscript received April 03, 2009; revised May 15, 2009. Current version published August 12, 2009.

M. Angelone, D. Lattanzi, and M. Pillon are with Associazione EURATOM-ENEA sulla Fusione, C.R. Frascati, C.P. 65-00044 Frascati, Rome, Italy (e-mail: maurizio.angelone@enea.it; angelone@frascati.enea.it; Daniele.Lattanzi@enea.it; Mario.Pillon@enea.it).

G. Aielli and R. Santonico are with the Dipartimento di Fisica, Università "Tor Vergata," 00133 Rome, Italy (e-mail: giulio.aielli@roma2.infn.it; Rinaldo.santonico@roma2.infn.it).

S. Almaviva, M. Marinelli, E. Milani, G. Prestopino, A. Tucciarone, C. Verona, and G. Verona-Rinati are with the Dipartimento di Ingegneria Meccanica, Università di Roma "Tor Vergata," I-00133 Rome, Italy (e-mail: Salvatore.Almaviva@enea.it; marco.marinelli@uniroma2.it; milani@ing.uniroma2.it; giuseppe.prestopino@uniroma2.it; aldo.tucciarone@uniroma2.it; claudio.verona@uniroma2.it; gianluca.verona.rinati@uniroma2.it).

R. Cardarelli is with Istituto Nazionale di Fisica Nucleare, "Tor Vergata," 00133 Rome, Italy (e-mail: roberto.cardarelli@uniroma2.infn.it).

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TNS.2009.2025177

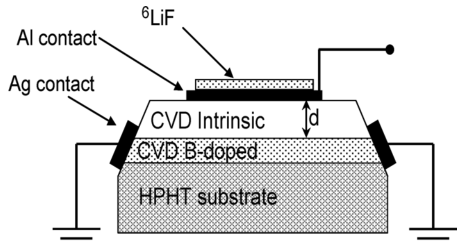

Fig. 1. Layered structure for the SCD detector.

any, with no need to mechanically remove it. The device is finally mounted in an housing [5].

The detector is usually biased with an electric field of about 2 $\mathrm{V}/\mu\mathrm{m}$ with positive polarity on the top electrode. At this stage, a standard characterization of the resulting device is performed by $^{241}\mathrm{Am}$ α particle detection. A 100% charge collection efficiency and energy resolution (FWHM) of approximately 1% are routinely observed. It ought to be stressed that the above results are well reproducible demonstrating that a standardization of the SCD performances is possible.

III. Diamond Detector Application

A. Thermal and Fast Neutron Detection

Fast neutrons are detected directly in the bulk of the intrinsic diamond layer through the $^{12}\mathrm{C}(\mathrm{n},\alpha)^{9}\mathrm{Be}$ and $^{12}\mathrm{C}(\mathrm{n},\mathrm{n}')^{12}\mathrm{C}*$ reactions [10]. The produced $^{9}\mathrm{Be}$ and α ions have a total energy $\mathrm{E}_{\alpha}+\mathrm{Be}=\mathrm{E}_{\mathrm{n}}-5.7\ \mathrm{MeV}$, where E_{n} is the energy of the impinging neutron. To detect both the fast and the thermal neutrons a layer of $^{6}\mathrm{LiF}$ or $^{10}\mathrm{B}$ which converts low energy neutrons into highly ionizing particles, is used.

To achieve sensitivity to thermal neutrons a $^{6}\mathrm{LiF}$ layer 95% enriched in $^{6}\mathrm{Li}$ was used first. $^{6}\mathrm{Li}$ interacts with thermal neutrons through the reaction $^{6}\mathrm{Li}(\mathrm{n},\alpha)\mathrm{T}$ producing 2.73 MeV Tritium and 2.07 MeV alpha particles which are easily detected by the diamond device. The $^{6}\mathrm{LiF}$ layer thickness is usually between 1 and 4 $\mu\mathrm{m}$, depending upon the required sensitivity. Fig. 2 reports the Pulse Height Spectrum (PHS) recorded locating the Lithium Diamond (LiDia) detector in a Polymethilmethacrylate (PMMA) phantom exposed to 14 MeV neutrons produced by the Frascati Neutron Generator [11]. The PMMA slows down a fraction of the fast neutrons allowing to get thermal neutrons. The two sharp peaks at about 2 and 2.7 MeV are due to α-particles and tritium ions produced by the $^{6}\mathrm{Li}(\mathrm{n},\alpha)\mathrm{T}$ reaction respectively. The width of these peaks (FWHM) depends upon the $^{6}\mathrm{LiF}$ layer thickness [12]. At higher energy (note the $\times 100$ amplified intensity scale on the right) the broader peak at about 9.1 MeV is due to the $^{12}\mathrm{C}(\mathrm{n},\alpha_{0})^{9}\mathrm{Be}$ reaction in diamond. This peak is well separated from the $^{12}\mathrm{C}(\mathrm{n},\mathrm{n}')^{12}\mathrm{C}* \rightarrow 3\alpha$ continuum at lower energies. The LiDia detector was also irradiated in a TRIGA thermal nuclear reactor in order to study its capability to withstand high thermal neutron fluxes (up to $2.0 * 10^{9}\ \mathrm{n}/\mathrm{cm}^{2}/\mathrm{s}$). The linearity of the device was also verified up to a counting rate of 150 KHz [7] limited by the slow electronics used. A test was also successfully performed to verify the capability of the detector to operate in current mode using a current amplifier.

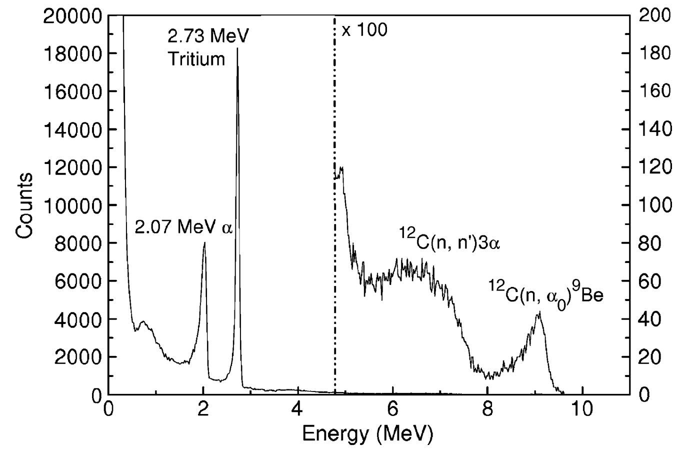

Fig. 2. A typical PHS measured with a LiDia Detector irradiated in a PMMA phantom with 14 MeV neutrons.

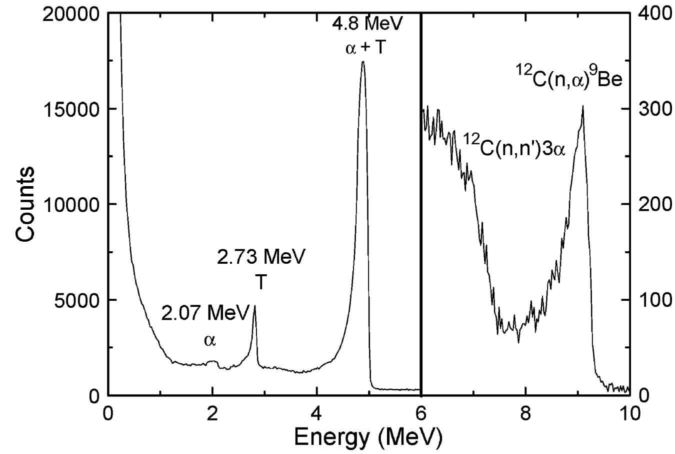

Fig. 3. PHS recorded with a $^{6}\mathrm{LiF}$ "sandwich" detector. The reaction with 14 MeV neutrons is evidenced in the right side.

B. Sandwich Detector

To increase the efficiency to thermal neutrons a new detector based on a sandwiched structure was produced. This detector is made with two layered detectors overlapped. The two intrinsic SCD films are facing each other and the thin layer of $^{6}\mathrm{LiF}$ is in between. From the kinematics of the $^{6}\mathrm{Li}(\mathrm{n},\alpha)\mathrm{T}$ reaction it is known that the reaction products (α and T) are emitted in opposite directions. The two reaction products are absorbed in the two intrinsic SCD diamond films which are connected in parallel mode. The PHS spectrum presents a single peak at an energy which is the sum of the kinetic energies of α and T ions (Fig. 3). Indeed, in Fig. 3 two residual 2.73 MeV and 2.06 MeV peaks are present (peaks integral about a factor 15 lower than the main one). These peaks are due to a slight misalignment of the two diamond detectors. Work is in progress to overcome this technical problem.

Once properly calibrated (e.g. in a standard thermal flux), the area under the main peak yields information on the thermal flux in which the detector is exposed or about the Li-6 reaction rate. The latter point has a direct application in fusion studies since

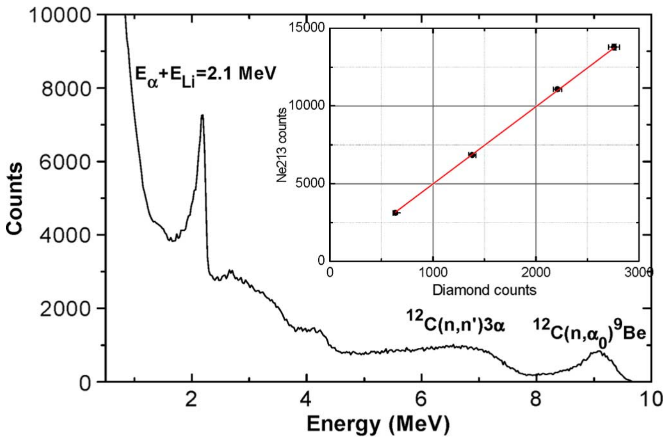

Fig. 4. a) PHS for Boron sandwich detector. The single peak is clearly evidenced. b) The inset shows the detector linearity tested versus NE-213 scintillator.

the next generation of fusion tokamaks will use the D-T nuclear reaction. T will be self produced in a region named breeding blanket (BB). The sandwich detector, once properly calibrated can be used to monitor both the total and time dependent tritium production in the BB. This is presently under testing and development, the main draw-backs being the very harsh working condition under which a tritium monitor will operate (high temperature and high neutron and gamma fluxes). Calibration can be performed also for the single layered detectors.

C. Boron Detector

A new detector using Boron as converting medium was recently realized. $\mathrm{B_2O_3}$ is evaporated on top of the conductive contact that in this case is made of Chromium. The boron layer thickness range from 200 to 750 nm. The $^{10}\mathrm{B(n,\alpha)^7Li}$ reaction is used in this case. $^7\mathrm{Li}$ can be either in a ground (6%) or excited state (94%). In the excited state the two reaction products carry away a total energy of $\mathrm{Q = 2.31\ MeV}$ ($^7\mathrm{Li} = 0.84\ \mathrm{MeV}$ and $\alpha = 1.47\ \mathrm{MeV}$) while in the ground state $\mathrm{Q = 2.79\ MeV}$ ($^7\mathrm{Li} = 1.015\ \mathrm{MeV}$ and $\alpha = 1.777\ \mathrm{MeV}$). Because of the low $^7\mathrm{Li}$ kinetic energy and of the energy absorbed in the Boron layer, in a PHS spectrum obtained irradiating the detector with thermal neutrons, it is not easy to discriminate the $^7\mathrm{Li}$ peak from the background.

To overcome this problem the "sandwich" structure has been realized and tested. The PHS spectrum is shown in Fig. 4. Its performances where tested at the 14 MeV Frascati Neutron Generator (FNG) [12] using a PMMA phantom and its linearity is shown in the inset of Fig. 4.

The Boron sandwich detector has a specific application in the field of boron neutron capture therapy (BNCT) [13]. This therapy uses thermal neutron irradiation of boron which is the element vehiculed inside cancer cells, where boron is preferentially absorbed. The short-range ions produced by the $^{10}\mathrm{B(n,\alpha)^7Li}$ nuclear reaction will deposit their energy in the cancer cells killing them. The boron-diamond sandwich detectors are very small, are made of carbon, which is tissue equivalent, and measure the $^{10}\mathrm{B(n,\alpha)^7Li}$ reaction. They represent the "optimal" neutron detector for such application since they can directly measure the energy deposited by the reaction products in the irradiated tissue.

Once properly calibrated in a thermal flux, by measuring the total $^{10}\mathrm{B(n,\alpha)^7Li}$ counts (area under the peak in Fig. 4) it is possible to determine the energy released in carbon (tissue). Alternatively, the boron-diamond sandwich detector can provide the actual thermal component at the irradiation position. Tests in a BNCT facility available at ENEA Casaccia laboratory are planned.

D. Neutron Detection in Tokamak

Artificial diamond detectors are successfully studied at JET tokamak since 2003 [8]. Three SCD detectors were installed at JET tokamak for the 2008 experimental campaign to measure total and time dependent neutron production during each D-D plasma pulse. The simultaneous measurement of total and 14 MeV neutron emission is possible. Two detectors (Det-1 and Det-3) made with 100 and 75 $\mu\mathrm{m}$ active diamond layer thickness respectively, are covered with 3 $\mu\mathrm{m}$ of $^6\mathrm{LiF}$ while the third one (Det-2, 200 $\mu\mathrm{m}$ thick) is covered with a layer (2 mm) of paraffin to enhance its response to DD (2.5 MeV) and DT (14 MeV) neutrons taking advantage of the high elastic cross section of neutrons with hydrogen (about 1 barn over the energy range of interest). The response of the diamond detectors is checked against the detectors routinely used at JET for total neutron emission (fission chambers) and 14 MeV neutron emission (silicon diodes). Det-1 and Det-2 operate using a charge preamplifier located inside the JET torus hall. To reduce the huge electromagnetic noise produced by the tokamak both detector and preamplifier are located in a metallic shielding box. The third detector (Det-3) is connected to a fast broad band preamplifier (DBA) located outside the torus hall through a high frequency, low impedance, low attenuation super-screened cable about 100 m long, and it is used to test new fast electronics (see next section).

Preliminary results for Det-1 and Det-2 are confirming the excellent performances of these detectors as it is indicated in Fig. 5. Low and high energy neutrons are discriminated using electronic thresholds. The three SCD detectors correlate very well with the JET neutron monitors (fission chambers and silicon diodes). The different slopes in Fig. 5 depends upon the different locations around the tokamak (the fission chamber is always the same) and the sensitivities (different volume) of the used detectors.

IV. Development of Fast Electronics for SCD

Two properties of a diamond detector to deal with are the already mentioned high band-gap value (5.3 eV) and the high carrier mobility. These properties led to signals having both lower amplitude (worse signal to noise ratio) and faster rising time (<100 ps) than that produced e.g. by a silicon detector. By experience, it is known that conventional charge preamplifiers coupled to diamond detectors yields excellent results in terms of energy resolution $(\mathrm{FWHM} < 1\%)$ and time stability (e.g. the results shown in Figs. 3 and 5). However, the charge preamplifiers have a long integration time (of the order of tens of $\mu\mathrm{sec}$) and they need to be located very close to the detector. This "traditional" electronics limits the applications of diamond detectors

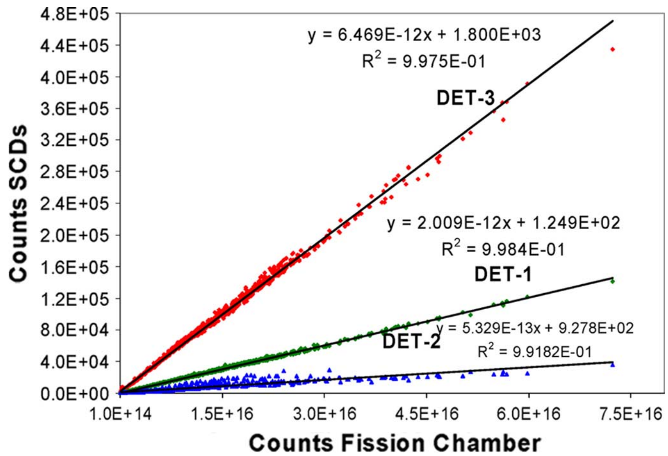

Fig. 5. Correlation among SCD detectors and Fission chamber at JET tokamak (results March–April 2008).

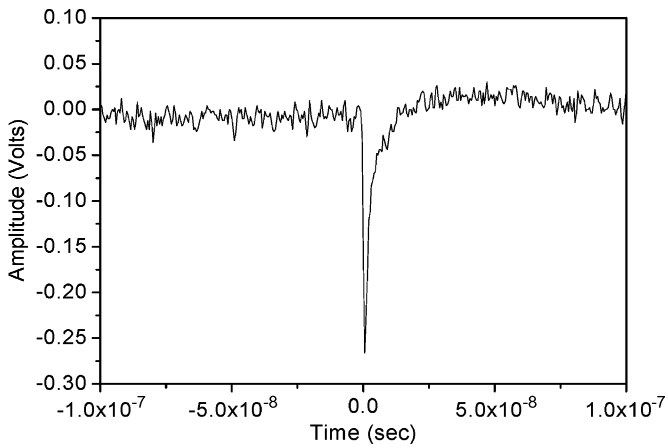

Fig. 6. Output pulse produced by a DBA preamplifier connected to a SCD diamond detector irradiated with 14 MeV neutrons.

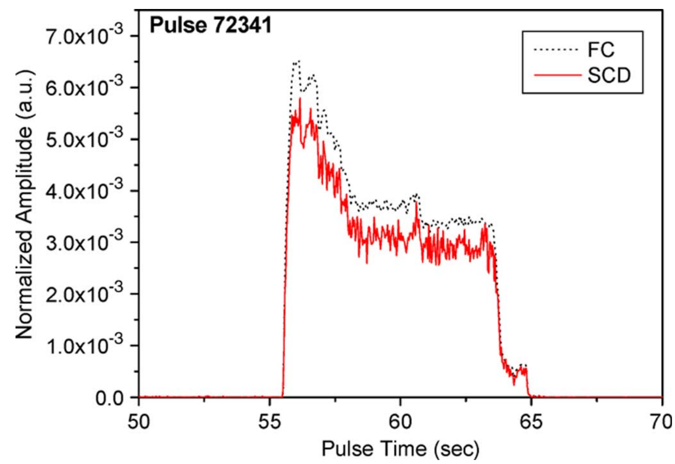

Fig. 7. Time dependent neutron emission during JET pulse #72341 recorded by Det-3 coupled to DBA and compared with one of the Fission Chambers routinely operating at JET.

to low count rates preventing their use in high neutron flux environments. The solution is in the remote processing of the small (some μV) and very fast $(\text{rising time} < 100\ \text{ps})$ signal produced in diamond. However, this is not an easy task. To avoid signal distortion and attenuation, high frequency, low attenuation, long super-screened cables are needed. These cables are of commercial type and must have a very low impedance at high frequency. Fast preamplifiers are located at the end of such long cable to boost the signal for the following acquisition which is performed using conventional electronics (threshold discriminator and counter).

This scheme is under testing at JET tokamak. Det-3 is connected to a low attenuation super-screened cable 100 m long. The pulses are sent to a diamond broad band (2 GHz) amplifier (DBA) [14]) and then to a threshold discriminator and a counter. The DBA was developed by GSI (Germany) for operating with diamond detectors and produces pulses with FWHM lower than 1 ns (Fig. 6) and allows amplification of electrical pulses from <1 mV up to 300 mV. Fig. 5 reports also the excellent correlation between the total neutron emission recorded by Det-3 and that measured with conventional fission chambers. Det-3 measures also the time dependent neutron emission, as shown in Fig. 7. Comparing the data it ought to be remembered that while the fission chamber is operating in current mode, Det-3 is working in pulse mode. The above results demonstrate the feasibility of this measuring scheme. However, these results do not fulfil all the requirements because the DBA does not allow to perform spectrometry. The latter is a major requests for the neutronics measurements in tokamaks because the neutron spectroscopy yields many physical information about the burning plasma (e.g plasma temperature).

The amplification process of DBA produces too short a pulse to be properly sampled with a commercial digitizer. This requires a further step which comprises the development of a conceptually new fast charge amplifier (FCA). In order to allow the diamond's output pulse to be sampled with a fast digitizer, the new FCA must amplify the charge produced in the detector by the ionizing radiation. The FCA must also stretch the small and very fast input signal produced by diamond, keeping proportional the output charge to the input one. This requires the input signal, which is of the order of tens of ps, to be stretched up to 50–200 ns. The output signal is thus sampled by the digitizer and the PHS is obtained via software analysis. The output signal must keep the information produced in the diamond detector and must not introduce noise and/or distortions that prevent its use for spectroscopy. A prototype of such preamplifier has been developed [15] in collaboration with the Department of Physics, of Rome "Tor Vergata" University and Istituto di Fisica Nucleare (INFN), "Tor Vergata" section. It is presently being tested and assessed and preliminary results are very encouraging.

The spectrometric performances of such device are shown in Fig. 8. This is the PHS spectrum of a three alpha peaks source $({}^{240}\mathrm{Pu}(5.3\ \mathrm{MeV}) + {}^{241}\mathrm{Am}(5.5\ \mathrm{MeV}) + {}^{244}\mathrm{Cm}(5.8\ \mathrm{MeV}))$ as recorded by a 2 GHz LeCroy digital oscilloscope. The peaks have an energy resolution (FWHM) of 1.5% since the satellite peaks are not (yet) resolved. The output pulse of the FCA is shown as well. The goal is to install one prototype of this fast stretching preamplifier at JET and to test it during the experimental campaigns.

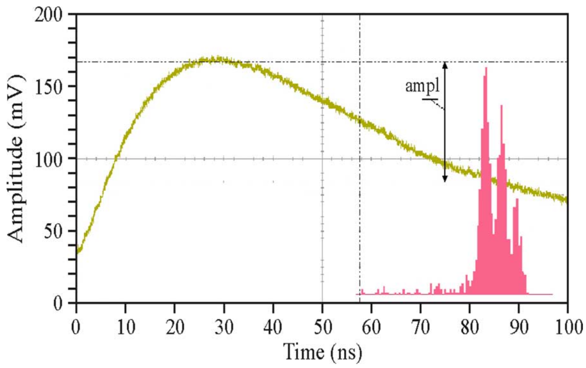

Fig. 8. PHS (histogram) as recorded by oscilloscope for the three alpha peaks source $(^{240}\mathrm{Pu}(5.3\ \mathrm{MeV})+{}^{241}\mathrm{Am}(5.5\ \mathrm{MeV})+{}^{244}\mathrm{Cm}(5.8\ \mathrm{MeV})$ by using the FCA developed in this work. The output pulse is also shown.

V. CONCLUSION

State-of-the-art artificial Single Crystal Diamond (SCD) neutron detectors based on a multilayered structure and grown by CVD technique have been discussed together with some relevant applications such as the simultaneous detection of fast and thermal neutrons possible thanks to the use of a thin $^6\mathrm{LiF}$ layer. The SCD properties, advantages and draw-back were also outlined as well as some possible new developments.

The SCD promises to be useful in radiation harsh environment such as those expected in a fusion tokamak. Tests are on-going at JET tokamak where advanced and innovative solutions such as the use of a SCD detector operating with a single cable and the fast charge preamplifier located far away, are under testing.

SCD detectors with layered structure and covered with boron where also developed and promise to be helpful dosimeters for the boron neutron capture therapy.

REFERENCES

[1] P. Bergonzo and R. Jackman, *Thin-Film Diamond II (Semiconductors and Semimetals 77)*, C. Nebel, Ed. New York: Academic, 2004, p. 197.

[2] A. Paoletti and A. Tucciarone, Eds., "The physics of diamond," in *Proc. Int. School of Physics "Enrico Fermi," Course CXXXV*, Varenna, Italy, Jul.-Aug. 1996, IOS Press 1997.

[3] R. J. Tapper, "Diamond detectors in particle physics," *Rep. Prog. Phys.*, vol. 63, p. 1273, 2000.

[4] W. Adam *et al.*, "Radiation hard diamond sensors for future tracking applications," *Nucl. Instrum. Methods Phys. Res. A*, vol. A565, p. 278, 2006.

[5] M. Marinelli, E. Milani, G. Prestopino, A. Scoccia, A. Tucciarone, G. Verona-Rinati, M. Angelone, and M. Pillon, "High performance 6LiF-diamond thermal neutron detectors," *Appl. Phys. Lett.*, vol. 89, p. 143509, 2006.

[6] A. Balducci, M. Marinelli, E. Milani, M. E. Morgada, A. Tucciarone, G. Verona-Rinati, M. Angelone, and M. Pillon, "Extreme ultraviolet single-crystal diamond detectors by chemical vapor deposition," *Appl. Phys. Lett.*, vol. 86, p. 193509, 2005.

[7] M. Marinelli, E. Milani, G. Prestopino, A. Tucciarone, C. Verona, G. Verona-Rinati, M. Angelone, D. Lattanzi, M. Pillon, R. Rosa, and E. Santoro, "Synthetic single crystal diamond as a fission reactor neutron flux monitor," *Appl. Phys. Lett.*, vol. 90, p. 183509, 2007.

[8] M. Angelone, M. Pillon, M. Marinelli, E. Milani, A. Paoletti, G. Pucella, A. Tucciarone, G. Verona-Rinati, L. Bertalot, F. Orsitto, S. Popovichev, and A. Murari, "Time dependent 14 MeV neutron measurement using polycrystalline CVD diamond detector at JET tokamak," *Rev. Sci. Instrum.*, vol. 76, p. 013506, 2005.

[9] N. Tartoni, M. Angelone, M. Pillon, S. Almaviva, M. Marinelli, E. Milani, G. Prestopino, A. Tucciarone, C. Verona, G. Verona-Rinati, and M. A. Roberts, "X-Ray detection by using CVD single crystal diamond detector," in *Proc. SORMA West*, Berkeley, CA, Jun. 2008.

[10] M. Pillon, M. Angelone, and A. Krasilnikov, "14 MeV neutron spectra measurements with 4% energy resolution using type IIa diamond detector," *Nucl. Instrum. Methods Phys. Res. B*, vol. B101, p. 473, 1995.

[11] M. Martone, M. Angelone, and M. Pillon, "The 14 MeV Frascati neutron generator," *J. Nucl. Materials*, vol. 212–215, p. 1661, 1994.

[12] S. Almaviva, M. Marinelli, E. Milani, G. Prestopino, A. Tucciarone, C. Verona, G. Verona-Rinati, M. Angelone, D. Lattanzi, M. Pillon, R. M. Montereali, and M. A. Vincenti, "Thermal and fast neutron detection in chemical vapor deposition single-crystal diamond detectors," *J. Appl. Phys.*, vol. 103, p. 054501, 2008.

[13] D. W. Nigg, "Recent trends and progress in the physics and biophysics of neutron capture therapy," *Prog. Nucl. Energy*, vol. 35, p. 79, 1999.

[14] P. Moritz *et al.*, "Diamond detectors with sub-nanosecond time resolution for heavy-ion spill diagnostics," in *Proc. 8th Beam Instrumentation Workshop, AIP Conf. 451*, Stanford, CA, 1998, GSI-RD42 Collaboration.

[15] M. Angelone, G. Aielli, S. Almaviva, D. Lattanzi, M. Marinelli, E. Milani, M. Pillon, G. Prestopino, R. Santonico, C. Verona, and G. Verona-Rinati, "Neutron spectrometry by means of artificial diamond detectors using a remote read-out scheme," presented at the ANIMMA Conf., Marseille, France, Jun. 7–10, 2009.