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POSITION-SENSITIVE SILICON DETECTOR FOR X-RAY DIFRACTOMETRY OF RAPID PROCESSES

The results of development and application of position sensitive microdetectors for studying dynamics of rapid processes in metals and alloys under heating/cooling by means of high-speed radiography have been presented.

Key words: position-sensitive detectors, TimePix silicon micro pixel detector, microstrip metal detectors, high-speed X-ray radiography, dynamics of phase transformations.

To form the properties of structural materials for micro- and nanotechnologies it is necessary to know the kinetics of rapid phase-structural transformations. The information about dynamics of rapid processes related to change in structural factors of samples exposed to heating/cooling is obtained by measuring shifts of diffraction peaks in angular distribution of X-rays scattered in the sample. The better are detector's positional sensitivity and time resolution, the higher is accuracy and the more detailed is information on dynamics of phase transformations. The study of phase transitions using the micropixel detector in picosecond range under complex irradiation of samples by laser and synchrotron radiation (Advanced Photon Source, USA) has been reported, for example, in [1]. The high-intensity X-rays required for the study of rapid processes complicate the problem of detector radiation hardness. All the known detector systems are characterized by significant factors of radiation impact on quality of their operation. The use of the next (fourth) generation X-ray sources (free electron lasers) having an intensity higher by 3—4 orders of magnitude opens new horizons for researching the fast processes (femtosecond level). However, it requires a new-type detector with extremely high radiation hardness.

As a part of basic research on high energy physics within the framework of budget-funded projects of the National Academy of Sciences of Ukraine (NASU), the Institute for Nuclear Research of NASU has created new-type detectors for measuring the spatial intensity distribution of charged particles and synchrotron radiation, i.e. the metal micro-detectors (MMD). In cooperation with MEDIPIX Collaboration (CERN) they has been used for the measurement of low-energy ion fluxes (laser mass spectrometer IAP of NASU) and synchrotron radiation (ESRF, Grenoble). The evaluative measurements have showed sufficient sensitivity of *TimePix* hybrid detectors (CERN) to study the dynamics of phase transitions in metals using a high-speed X-ray installation (IPM of NASU).

This paper presents the first results of development and application of position-sensitive X-ray micro-detector as an element of experimental techniques for study of rapid processes of phase transitions in metals and their compounds during heating/cooling or compression/tensile. The results were obtained using micro-strip and micropixel detectors as prototypes, including those on the basis of radiation-resistant metal micro-sensors created on the basis of original technique in the INR NASU [2]. The research was conducted at the upgraded fast X-ray diffraction facility of the Institute for Problems of Materials Science of NASU [3] using a *TimePix* silicon micropixel detector [4].

EXPERIMENTAL METHOD

The effective application of silicon position-sensitive detectors designed for the purposes of experimental high energy physics, namely, for recording low-energy ions [4] and synchrotron radiation [5] was an impetus for the development of position-sensitive micro-detector. Addressing the problem of sensor radiation hardness the INR of NASU in cooperation with MEDIPIX Collaboration (CERN) and IAP of NASU has been developed and tested on charged particle beams and synchrotron radiation the prototypes of metal micropixel and micro-strip detectors which are resistant to radiation by definition of their physical and technical principles [6].

The use of micro-detectors in high-throughput X-ray analytical facilities makes it possible to remove the shortcomings of mechanical scanning, since the angular distribution of X-rays is measured by individual micro-sensors (with a typical size of about 50 microns) immediately and simultaneously in the whole geometric acceptance of detector. Actually, here, there is implemented the so-called electronic photo-plate mode with possibility to display digitally the diffraction peak position as a function of various physical factors (time, temperature, pressure, etc.). This allows the researchers to get more information received about the stages and mechanisms of phase and structural transformations through appropriate analysis of diffraction pattern evolution. The high-throughput X-ray diffraction facility (Fig. 1) with the use of which the study has been conducted is described in [3].

RESEARCH RESULTS

The *TimePix* hybrid micropixel silicon detector (MEDIPIX Collaboration (CERN, Geneva)) is described in detail in [4]. It consists of 300-micron layer of silicon semiconductor sensor connected by indium impregnations («bump-bonding» technique) with micropixel chip reader made for industrial CMOS-0.25 micron technology in six metallic layers. The geometry of both parts is the same: a matrix (256 × 256) pixels with each having an area of (55 × 55) μm^2 (Fig. 2). This means that the detector's sensitive area is equal to (14 × 14) mm^2. Each pixel of microchip reader (based on more than 400 transistors) has pre-amplifier, discriminator, and counter (the maximum amount of events is 11 840). Due to an extremely low capacitance at the entrance of amplifier (~1 fF) ensured by «bump-bonding» technique the equivalent noise of each channel does not exceed 750 electrons. When the X-ray quantum (energy 10 keV) falls on the sensor layer, the microchip generates a signal in the form of the number of events proportional to amplitude by time-over threshold (ToT) principle.

The data were read-out to PC via USB-port using a new *FitPix* interface device and PIXELMAN software created by the Institute for Experimental and Applied Physics (Prague) [8]. The new interface allowed the researchers to achieve exposures with frequency of up to 100 Hz. The study was performed at exposure from several dozen to several hundred milliseconds. The geometric dimensions of detector sensitive surface providing a spatial accuracy of diffraction maximum position ~20 μm covered an angle of 14° (4θ by Seeman-Bohlin) with an angular resolution of 0.017°. *TimePix* is a sort of electronic «photo film» capable of displaying real-time dynamics of rapid transformations.

The diffraction peak position was determined on the basis of two-dimensional intensity distribution of X-rays scattered on metal sample obtained with the help of *TimePix*. The sample was heated at a rate of $V_h \approx 250$ °C/s by passing an electric current through it and was cooled in a vacuum at a rate of $V_c \approx 50$ °C /s within the temperature range from 20 to 1500 °C. The data discreteness was set by time exposures in *TimePix*,

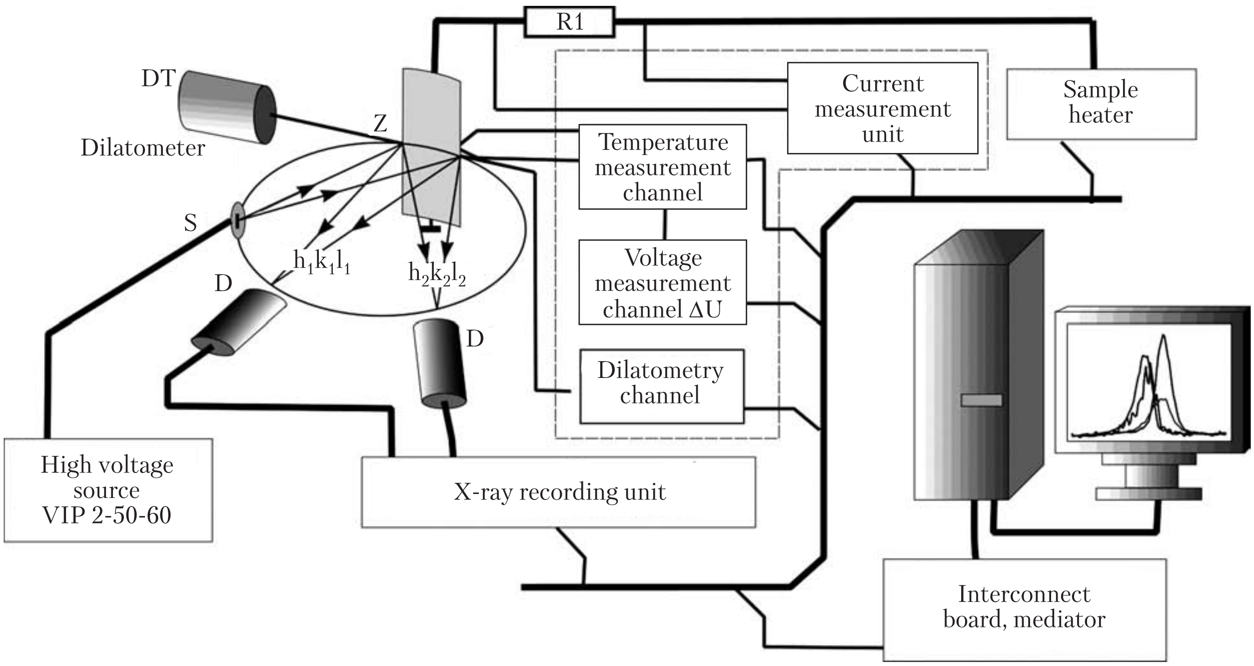

Fig. 1. Flowchart of fast X-ray diffraction facility: S is focus of X-ray source, Z is studied metallic sample, DT is dilatometer, D is X-ray micro-detector

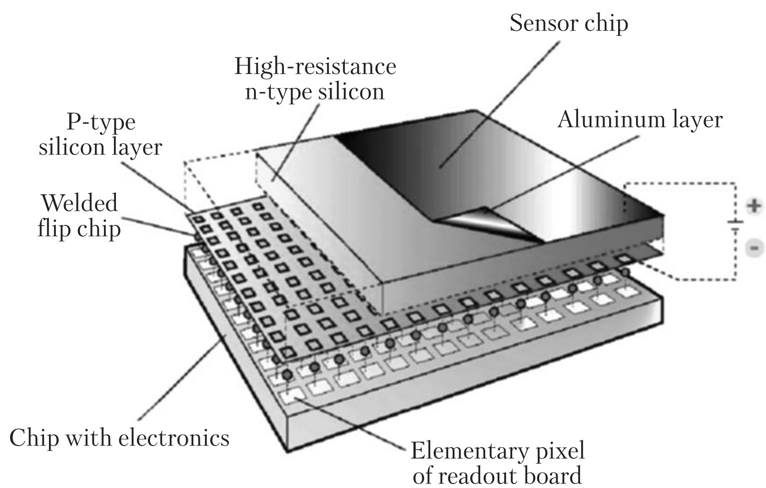

Fig. 2. Schematic representation of TimePix hybrid detector. The upper part: semiconductor silicon sensor with (256 × 256) micropixel structure made of high-resistance n-type silicon; the lower part: (256 × 256 channels) microchip reader made of low-Ohm silicon and connected to sensor matrix

which in different experiments varied from a few dozen to a few hundred milliseconds. For the complete measurement cycle, during 10—20 seconds, this meant the accumulation of several hundred numerical (256 × 256) matrices, with the number of events in each of the 65 536 cells corresponding to the intensity of quanta recorded. The quantum energy was equal to 10 keV.

Fig. 3 shows the distribution of intensity of X-rays scattered on metallic sample measured in one of exposures by *TimePix* silicon detector. Each exposure corresponds to a certain time after

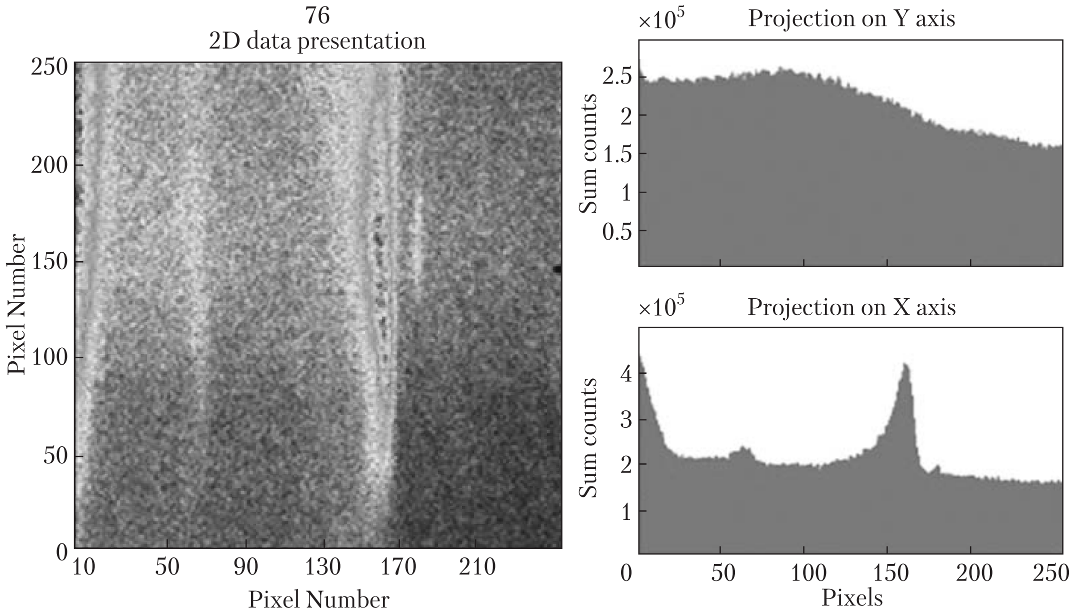

Рис. 3. On the left: intensity distribution of X-rays scattered on metal sample measured by *TimePix* silicon detector in high-throughput X-ray diffraction facility. On the right: projection of this two-dimensional distribution on the Y and X axes

the start of experiment, which runs a system of gradual increase in electric current through the sample and, therefore, heats the sample. Time (in seconds after the start of experiment) is marked with capital letters (76) over the two-dimensional distributions («2D data presentation»), where the X- and Y-axes have 256 gradations according to the number of pixels in *TimePix* detector. The intensity of X-rays registered in pixels by *TimePix* detector is displayed by color (blue color for low intensity, the number of events from 0 to 5; red color for high intensity, the number of events from 20 to 25) or by corresponding gradation of dark-white proportion. To obtain information about the evolution of diffraction peak position depending on temperature the two-dimensional distributions were projected on the X axis (the bottom right picture on each frame).

The sample temperature varied within the range from 20 to 1250 °C. The angular position of diffraction peak for sample's alpha phase changed from 21.5 to 24.7°, with its total intensity (net of background interpolated from outside of diffraction peak to its area) decreasing almost 15 times reflecting the virtual disappearance of alpha phase in heated sample.

The detailed data were obtained using a special software for *Armco* iron sample whose chemical composition corresponds to the annealed steel ARMCO/E12 composition (content of copper, manganese, and silicon does not exceed 0.3%, that of sulfur, phosphorus, and carbon is less than 0.025%). These data are presented in the form of temperature dependence of alpha- and gamma-phase parameter within the temperature range from 20 to 1250 °C (Fig. 4). The initial stages of transformation were accurately recorded on the basis of changing intensity of alpha phase transformed, whereas the final stage was monitored by changing intensity of gamma phase formed. The high time resolution of method for fast X-ray diffraction allowed the researches to determine the time interval of alpha-gamma transformation with sufficient accuracy (~0.05 s) at a heating rate of ~250 °C/s and to build the kinetic curves of transformation (see Fig. 4). The accuracy of data obtained for alpha and gamma parameters is 1.2×10^{-3} Angstrom units.

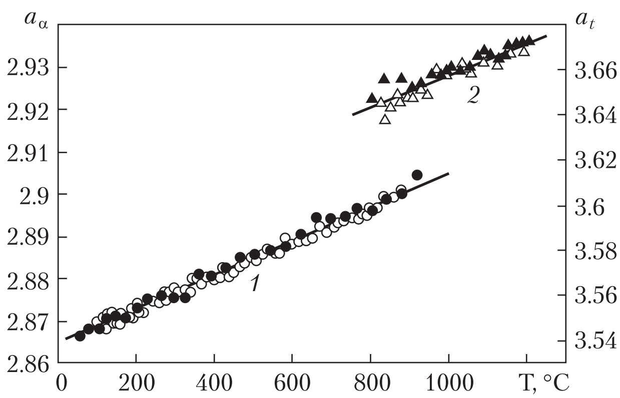

Рис. 4. Parameter of alpha (ferrite) and gamma (austenite) phases of sample's crystal lattice depending on sample temperature. Dark symbols: data obtained for the sample when heated; light symbols: data obtained the sample when cooled

The advantages of *TimePix* detector are high resolution of two-dimensional distributions (pixel size is 55 × 55 μm^2) and possibility to observe the results of measurements in real time (with a frequency of 100 Hz). The shortcomings of device are limited size of detector (14 × 14) mm^2 and high cost of detector modules.

The results of recent studies of one-dimensional detector prototypes have proved good prospects for the development of cost-effective wide-range position-sensitive detector system for studying the dynamics of phase transitions on the basis of one-dimensional silicon micro-strip detectors [9] and XDAS commercial data readout system [10]. Three 128-channel detector modules are planned to be manufactured and installed in focal plane of device for the purpose of X-ray diffraction study of rapid processes in IPM NAS of Ukraine.

CONCLUSIONS

The evolution of positions of scattered X-rays diffraction maximums with positional accuracy of approximately 20 microns was observed while the samples were heated or cooled in the installation for high-throughput X-ray diffraction analysis using *TimePix* silicon micropixel detector. This has allowed the researchers to determine the parameters of alpha and gamma phases in Armco iron within the temperature range from 20 to 1200 °C during the transition from ferrite to austenite with accuracy of ± 1.2 × 10^{-3} Angstrom units. The data on phase transitions in metals (Fe, Cr, Ti, Zr, and Hf) and new samples of micropowder alloys have been obtained. The devices and software based on data readout commercial systems for multi-channel micro-strip detectors have been designed and developed. They constitute the basis for the development of wide-range position-sensitive detector system for ensuring a new level of studying the rapid processes and properties of materials with the use of X-ray diffraction on the samples based on cutting-edge micro- and nanotechnologies.

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В.М. Пугач

ПОЗИЦИОННО-ЧУВСТВИТЕЛЬНЫЙ КРЕМНИЕВЫЙ ДЕТЕКТОР ДЛЯ РЕНТГЕНОВСКОЙ ДИФРАКТОМЕТРИИ БЫСТРОТЕЧНЫХ ПРОЦЕССОВ

Представлены результаты разработки и применения позиционно-чувствительных микродетекторов для исследований методом скоростного рентгенографирования динамики быстротечных процессов в металлах и их сплавах при нагревании/охлаждении.

Ключевые слова: позиционно-чувствительные детекторы, микропиксельный кремниевый детектор TimePix, микростриповые металлические детекторы, скоростное рентгенографирование, динамика фазовых преобразований.

В.М. Пугач

ПОЗИЦІЙНО-ЧУТЛИВИЙ КРЕМНІЄВИЙ ДЕТЕКТОР ДЛЯ РЕНТГЕНІВСЬКОЇ ДИФРАКТОМЕТРІЇ ШВИДКОПЛИННИХ ПРОЦЕСІВ

Наведено результати розробки та застосування позиційно-чутливих мікродетекторів для досліджень методом швидкісного рентгенографування динаміки швидкоплинних процесів в металах та їх сплавах при нагріванні/охолодженні.

Ключові слова: позиційно-чутливі детектори, мікропіксельний кремнієвий детектор TimePix, мікростріпові металеві детектори, швидкісне рентгенографування, динаміка фазових перетворень.

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