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Bent crystals obtained by low energy plasma enhanced chemical vapour deposition for medical applications

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Summary. — This paper presents bent crystals obtained by low energy plasma enhanced chemical vapour deposition technique, which could be further improved for medical applications to focus gamma rays. Medical imaging plays an important role in the field of nuclear medicine and the use of bent crystals to focus energetic gamma rays used for imaging of radioactivity in the human body is our interest. An innovative technique to obtain curved crystals is proposed in this work. Surface profilometry measurements were done to find the curvature of curved crystals.

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1. – Introduction

Radioactive materials used as a diagnostic tool can identify the status of a disease and minimize the need for surgery by reducing the risks from postoperative infection. Nuclear imaging is performed by injecting a radiopharmaceutical into the patient and measuring the intensity distribution of gamma radiation emitted from the patient's body [1].

In order to have a good reflection efficiency and broad energy band-pass, special crystals like mosaic crystals and bent crystals with the controlled lattice deformation are needed. Curved crystals are promising because their curved diffracting planes allow focusing and concentrating gamma rays with higher diffraction efficiency in comparison with mosaic crystals leading to increasing energy bandwidth due to their continuum angular spread [2,3]. Bent crystals are nowadays largely used to excite coherent interactions of high-energy particles in beam collimation and also in beam extraction [4, 5].

The development of bent crystals in order to focus gamma rays will find significant application in the field of medical imaging. Short-lived radioisotopes are preferred for use in these tracers to minimize the radiation dose to the patient. In most cases, these

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radioisotopes decay to stable elements within minutes, hours, or days, allowing patients to be released from the hospital in a relatively short time. The radioisotope used in about 80 percent of nuclear diagnostic procedures is Tc-99m. The penetrating properties of its gamma rays and its short half-life (6-hours) help in reducing risk to the patient from more prolonged radiation exposure. Short-lived radio-nuclides such as technetium-99m, gallium-67, and thallium-201 are often used to diagnose the functioning of the heart, brain, lung, kidney or liver.

Cancerous cells have high growth rate and multiply very rapidly. The radioactive isotope injected into the body of the patient normally migrates to high growth rate locations and will incorporate in this new growth and if the injected substance is a short-lived radioactive isotope, the tumour location can be identified from the region of high radioactivity.

2. – General background

The diagnostics is based on Bragg diffraction of gamma rays from crystal planes. Diffracting crystals (Bent crystals) are used for focusing the gamma radiation and directing the radiation to a detector which could be used to analyze the data with respect to the location of the radiation source.

2˙1. *Crystal diffraction.* – Diffraction inside the crystal can either occur near the surface, *i.e.* Bragg geometry or in the volume, where the beam propagates through the entire crystal, namely Laue geometry. In the Laue mode, the incident beam of radiation enters from one surface of the diffracting bent crystal and interacts with a particular array of parallel atomic layers. After that, the radiation splits into transmitted and diffracted beam produced by a coherent superposition of scattering by the atoms in the parallel atomic layers. In the Bragg mode, the diffracted beam (which consists of almost all the incident energy) is produced by the multiple scattering with the atoms in the particular array of parallel atomic layers and the diffracted beam exits from the same surface where the radiation entered. For instance, bent crystals in Bragg geometry are currently used as Goebel mirrors to transform a divergent X-ray beam into an intense parallel beam.

It is well known demonstrated that gamma rays interact coherently in a crystal lattice if and only if the angles of the incident photons are very small and satisfy the Bragg relation and the crystal diffraction of gamma rays is given by

$$\sin\theta = \left(\frac{n\Lambda}{2d_{hkl}}\right), \tag{1}$$

where Λ is the wavelength of the gamma ray in Å, n is the order of diffraction, d_{hkl} is the d spacing of the planes hkl and θ is the crystal diffraction angle.

$$\Lambda = \left(\frac{hc}{E}\right), \tag{2}$$

where E is the gamma-ray energy (in keV), h is Planck's constant and c is the speed of light.

The approximate focal length (in metres) of a Laue-based lens is given as

$$F = 1.65 \cdot 10^{-2} \cdot E. \tag{3}$$

Smither [6] calculated the distance from the source to lens (L_S) and the distance from the lens to detector (L_D) and also the values of L_S and L_D for different gamma-ray sources were tabulated using the formula

$$\frac{1}{F} = \frac{1}{L_S} + \frac{1}{L_D}\,, \tag{4}$$

In his experiment, for a 100 keV gamma ray diffracted by the (111) planes of germanium, θ is found to be 1.0895 degrees or 0.01902 rad and hence the surface of a Bragg diffraction crystal has to be 52.6 times longer than the height of a Laue crystal used in diffracting the same beam. As reported by Smither, for energetic gamma rays the diffraction angle (Bragg angle) is quite small and Laue geometry makes much more efficient use of the crystalline material.

Bending of the crystal eliminates the spreading of the diffracted beam in the plane perpendicular to the diffraction beam. Roa [1] used copper crystals due to their high reflectivity for gamma-ray energies of 100–200 keV. Halloin [7] explained that the use of perfect monocrystals for gamma-ray lens is not applicable due to the narrow diffracting angular range and also the thickness of a perfect crystal is greater than the extinction length. This latter leads to the formation of the constructive and destructive interferences limiting the diffracting power of crystal.

2.2. *Laue lens for medical application.* – A Laue lens is generally made of crystals which are arranged in concentric rings and oriented in such a way to focus X-rays or gamma rays through Bragg diffraction in Laue geometry. Laue lens is an emerging technology to realize the space-borne telescopes which are more sensitive within 100 keV–1.5 MeV and the concentration of gamma rays has been studied [8]. Development of Laue lens in focusing gamma rays in medical application finds a new way in the field of nuclear imaging. Smither [9] in his new experimental approach used crystals with curved diffracting planes including Si crystals and Si-rich SiGe gradient crystals to diffract high energy X-ray from advanced photon source and gamma rays from radioactive source and it was found that with the help of this approach one can increase the diffraction efficiency and bandwidth by a factor of 5 compared to mosaic crystals.

2.3. *Experimental layout for medical imaging.* – Diagnostic techniques in nuclear medicine use radioactive tracers which emit gamma rays from the inside of the body. These tracers are generally short-lived isotopes linked to chemical compounds which permit specific physiological processes to be scrutinized. They can be imparted to a patient by injection, inhalation or orally. The first type is where single photons are detected by a gamma camera which can view organs from many different angles. The camera builds up an image from the points from which radiation is emitted; this image is enhanced by a computer and displayed on a monitor for indications of abnormal conditions.

Bent crystals are expected to have both high diffraction efficiency and relatively broad energy bandwidth and would be capable of detecting the radiopharmaceuticals that emit 50–200 keV gamma rays which are generally used in diagnostic molecular imaging and in nuclear medicine. This system will focus the incoming gamma rays into a small focal point on a detector's sensitive area.

Figure 1 shows the experimental arrangement of the treatment method. A common method to locate cancerous tissue in the human body is to inject a biological compound that is readily incorporated in cancer cells and can carry a radioactive nucleus with

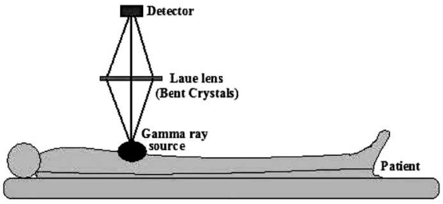

Fig. 1. – Pictorial representation of the clinical treatment method.

it, which is also incorporated in the cancerous cells. We are planning to carry out an experiment with these bent crystals so that they can be used in medical imaging.

3. – Fabrication of bent crystals by LEPECVD

Bent crystals can be obtained in different methods like by mechanically bending a perfect single crystal [6], by using mixed crystals with a composition gradient [10], by applying a thermal gradient to the planes of a perfect crystal [6] and by indentation method [2, 11]. In this present work bent crystals are obtained by low energy plasma enhanced chemical vapour deposition (LEPECVD) technique. The significant features of this technique are wide range of epitaxial growth rate at low substrate temperature and high reproducibility [12].

With the use of LEPECVD, a crystalline germanium film is grown on the surface of the silicon substrate (100) which is tilted 6° towards $\langle 111 \rangle$ and/or vice versa. The thickness of the silicon substrate is $400\,\mu$m. The sample details are given in table I. The importance of this method is to produce intrinsically curved crystals without the use of any mechanical means. The sample gets curved due to the lattice mismatch between Si and Ge and the different thermal expansion coefficients [13]. Thanks to this technique, curved crystals can be obtained by depositing tensile or compressive films over bulky substrates. This technique is chosen for its reproducibility, its capability to bend the crystal to the desired curvature, bending is strong due to high stress imparted which can be matched with stability without delamination and also the interjunction stress is distributed over a range of distances.

Table I. – *Details of the samples with the different thickness of Ge.*

Sample no.	Film thickness (nm)
1	505
2	762
3	1122
4	1800

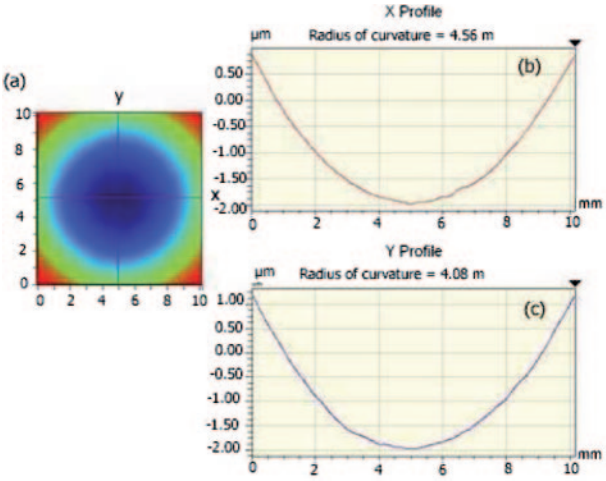

Fig. 2. – Optical surface profilometry of sample 4. (a) gives the false-colour representation of the deformation. The deformation pattern cross-section along the x (b) and y (c) directions is seen with the two curvatures. The isotropic bending has been achieved.

4. – Preliminary results and discussion-surface profilometry measurements

Wyko surface profiler systems are non-contact optical profilers to measure a wide range of surface heights. The basic principle used here is that the light reflected from a reference mirror combines with the light reflected from a sample to produce interference fringes, where the best-contrast fringe occurs at best focus. We used the vertical scanning interferometry mode in which the white-light source is filtered with a neutral density filter, which preserves the short coherence length of the white light, and the system measures the degree of fringe modulation, or coherence, instead of the phase of the interference fringes. The radius of curvature is measured with the help of surface profilers Wyko NT1100.

As an example, the optical surface profilometry of sample 4 is shown in fig. 2. The curvature has a significant effect on the diffraction efficiency of the crystal. The origin of the curvature can be interpreted in terms of the thickness of deposition and also due to lattice mismatch. The bent crystal samples of different thickness of germanium over the silicon substrate were grown and characterized. As the thickness of deposition of Ge is increased, the radius of curvature is decreased. Table II shows the variation of the curvature radius along the y direction with respect to the different thickness of Ge. The

Table II. – *Variation of the curvature radius with respect to the different thickness.*

Sample no.	Radius of curvature (m)
1	24.68
2	10.83
3	9.21
4	4.08

significance of this study is that it is easy to tailor the curvature to a wanted curvature after deposition of a suitable thickness and highly bent crystals can be produced by such a technique.

All the samples have been produced and optically characterized at Sensor and Semiconductor Laboratory (Ferrara, Italy).

5. – Simulation to optimize crystal diffraction properties

Performance of Laue medical lens relies on crystals as its most important elements. Hence, in order to maximize diffraction properties of the lens, crystal parameters have to be optimized. For this aim, a simulation code has been developed. This has been worked out through Python high-level programming language, and it is specifically designed for bent crystals. Indeed, the software describes diffraction in crystals with curved diffracting planes and generates the physical quantities which are typically used to characterize the diffraction properties of a crystal. These are diffraction efficiency and reflectivity as defined in [8]. The former is the ratio of the diffracted beam intensity over the transmitted one when no diffraction condition occurs and the latter is the ratio of the diffracted beam intensity to the incident beam intensity. Reflectivity can be determined by simply multiplying efficiency to the attenuation factor due to linear absorption in the sample. Moreover, for a given atomic number and reflection plane, the software code computes the thickness maximizing reflectivity and efficiency as a function of photon energy and angular spread, this latter being the bending angle of the crystal. Crystal thickness is a crucial parameter for optimization of a Laue lens. In Laue geometry, this is considered as the part of the crystal which is totally traversed by the radiation, giving the contribution to photon diffraction. In medical imaging applications of Laue lens, the thickness of crystal element gives the spatial resolution of the whole lens. Thus, this feature has to be taken into account when maximizing reflectivity and diffraction efficiency of the crystals composing the lens.

As a result of simulations, 5 mm thickness of sample 1 would diffract 140.5 keV of Tc-99m featuring 83% diffraction efficiency (which turns into a reflectivity of 42%), over 42 arcsec angular spread. Crystalline planes of Ge have been considered for diffraction. Hence, provided that the crystalline planes of the sample are curved, high-efficiency diffraction of gamma rays would be attained, with higher spatial resolution than for currently operating devices.

6. – Conclusions

The curved crystals obtained by LEPECVD could be used to focus and concentrate gamma rays coming from the body of the patient towards the detector. This work relates to a method of medical imaging and a Laue lens made of bent crystals would act as a device to efficiently focus gamma radiation. The basic application of crystal diffraction in focusing the energetic gamma rays has been explained. Thanks to these crystals, high spatial resolution could be attained, even preventing the patient from heavy doses of radioactivity, can enhance the overall performance of focusing the gamma rays and can also eliminate the filters provided by gamma camera.

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