Colloquia: Channeling 2010

Grown and artificial mosaic GaAs crystals for hard X-ray astronomy

E. Buffagni(*), C. Ferrari, L. Zanotti and A. Zappettini

Istituto IMEM-CNR - Parco Area delle Scienze 37/A, 43124 Parma, Italy

Summary. — In order to increase the collection efficiency of a lens for hard X-ray energies, mosaic GaAs crystals to be used as optic elements in Laue diffraction are proposed. In fact, GaAs crystals show a natural degree of mosaicity due to the spontaneous formation during the growth of cellular structures with dislocations at the boundaries. Several GaAs samples grown by LEC method have been characterized by means of high-resolution X-ray diffraction. Mosaicity values ranging from 10 to 25 arcsec have been measured. Since proper growth conditions allow to control and modify both the dislocation density and the cellular structure responsible of the mosaic spread, the possibility of obtaining crystals with a given degree of mosaicity by tuning the LEC growth conditions is proposed. A complementary strategy to increase the Darwin width of the diffraction curve based on curved crystals has also been proposed. The lattice curvature was achieved by introducing a compressive stress on the crystal surface. Curvature radii between 2 and 8 m were easily obtained in wafer crystals 500 μm thick.

PACS `95.55.Ka` – X- and γ-ray telescopes and instrumentation.
PACS `61.05.cp` – X-ray diffraction.
PACS `81.05.Ea` – III-V semiconductors.

1. – Introduction

The use of a Laue lens for space astronomy to focus hard X-rays with energies $E > 70$ keV seems a promising approach [1-8]. A Laue lens is a system constituted by a wide number of crystals with lattice planes oriented perpendicularly to the lens surface, so that Bragg diffraction in transmission configuration can be exploited. The focused beam should converge on a detector in the focus of the lens. The requested angular acceptance for this kind of system is of the order of a few tens of arcseconds, depending on the focal length. Owing to this, the use of perfect crystals as optical elements is inefficient due to their very narrow angular diffraction range for high energetic gamma-rays [9]. For example, in the case of the Ge 400 diffraction at an energy of 500 keV, the full width at

---

(*) E-mail: `elisa.buffagni@imem.cnr.it`

half maximum (FWHM) of the diffraction peak is only 0.06 arcsec. In order to increase the collection efficiency, mosaic crystals are proposed with mosaicity σ of the order of a few tens of arcseconds [1, 3-8]: the diffracted energy bandwidth at the Bragg angle θ_B that we can obtain is $\Delta E = E\sigma/\tan\theta_B$. More recently bent crystals have been also considered, where the curvature can be obtained with thermal gradient or composition graded crystals [1, 2, 7, 10] or indented crystals [11].

The choice of the material to be used as a diffracting element in a Laue lens is a trade-off among diffraction efficiency, absorption, and availability of crystalline material at sustainable cost. The diffraction efficiency of a crystal is, as a first approximation, proportional to its electronic density [4]. Thus, among the materials with high electron density, those that can be grown with proper quality and mosaicity (depending on the Laue lens peculiarities) should be chosen. Crystals of copper [3, 8], germanium [6, 12-15] and silicon-germanium [10] alloys have already been employed as diffracting materials for Laue lens, but a critical point remains the control of the mosaicity, particularly at low values.

Gallium arsenide (GaAs) has the same crystal structure (fcc), lattice spacing, and electron density of germanium. Moreover, the spontaneous formation of "cellular structures" with dislocation distribution at the boundaries between perfect zones of the crystal [12] can favour the growth of crystals with moderate degree of mosaicity as required by a Laue lens for hard X-rays.

In the present work we first consider the use of GaAs crystals as optical elements for hard X-ray astronomy. We present some results of high resolution X-ray diffraction characterization of GaAs crystals grown in different conditions by liquid encapsulated Czochralski method. Thus, we propose a complementary strategy to increase the Darwin width of the diffraction curve: the use of "artificial mosaic" crystals by bending crystals [16]. More in details, we propose a Laue lens optical element constituted by a stack of curved crystals.

2. – Experimental

The GaAs crystals were grown by the Liquid Encapsulated Czochralski (LEC) technique [17] in a high-pressure puller (LPAI Galaxie Model) with the configuration schematized in fig. 1a. The main growth parameters, common to all runs, are the B_2O_3 thickness (15 mm), the Ar pressure (20 atm), the thermal gradient at the solid/liquid interface (in the range 30–50 °C), the pulling rate (10 mm/h), and the growth direction $\langle 100 \rangle$ (see fig. 1a). The starting materials were pre-synthesized directly in the puller from different charges: Cr (sample LPA96) or Si (samples Si107-32 and 88-47) doped, As-rich (samples LPA190 and 191), or stoichiometric undoped (samples LPA183, 193, 206 and MB). All the samples, with the exception of Si9822-37 that is a commercial GaAs wafer doped with silicon, are slices cut from the ingots perpendicularly to the growth direction; a subsequent chemical etching with a HCl/HNO$_3$ 1:1 solution removed a thickness of about 50 μm where cut damaging occurred.

High-resolution X-ray diffraction measurements were performed by means of a X'Pert PRO Philips diffractometer, where the Cu$K\alpha_1$ radiation ($\lambda = 0.15405$ nm) is selected by four 220 reflections in a germanium monochromator.

Double-crystal X-ray topographs have been obtained in a RTK2 diffractometer using a perfect, dislocation-free, asymmetric cut Ge 620 monochromator. In this system, a perfect Ge or GaAs crystal shows a rocking curve with FWHM of 9 arcsec. The topograps have been taken with incident radiation at 80% of the reflectivity peak using the Cu$K\alpha$ 620 reflection geometry.
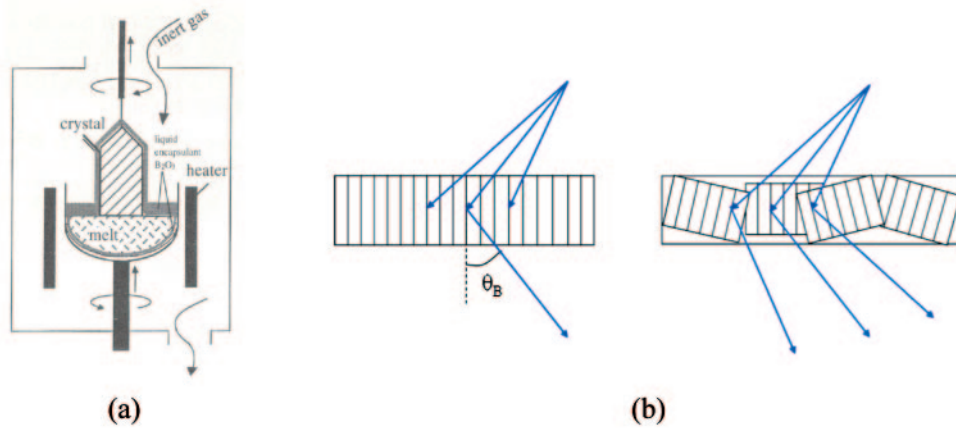
Fig. 1. – (a) Scheme of the LEC growth system. (b) Schematic comparison between a perfect crystal (on the left) and a mosaic one (on the right). The Bragg angle is indicated in the case of radiation impinging on a perfect crystal.

3. – Mosaic crystals: basics of crystal growth

A mosaic crystal, as firstly stated by Darwin [18], is a crystal made of microscopic nearly perfect domains, called crystallites, slightly disoriented with each other (fig. 1b). The distribution function of the misalignment ϑ with respect to the mean direction can be expressed by the Gaussian distribution

$$(1) \qquad W(\vartheta) = \frac{1}{\eta\sqrt{2\pi}} \exp\left[-\frac{\vartheta^2}{2\eta^2}\right].$$

The full width at half maximum (FWHM) of this distribution defines the mosaic spread, or mosaicity, σ of the crystal: $\sigma = 2\eta\sqrt{\ln 4}$; it is the average misalignment of the crystal domains.

GaAs crystals are usually characterised by the presence of "cellular structures" with dislocations distributed at the boundaries between perfect zones. The dislocation distribution gives rise to a real mosaic crystal with crystallites defined by the cell, and the dislocation density determines the disorientation between adjacent cells [19]. As an example, double-crystal topography of an undoped GaAs crystal is reported in fig. 2 where the different shades of grey evidence slight local misalignments between adjacent crystallites.

The cellular structure, and therefore the mosaic spread, can be controlled during the growth procedure.

The origin of the cellular structure is attributed to the effect of constitutional supercooling [20]. During the crystal growth from the melt (this is the case, for example, of the Czochralski method employed to grow the crystals presented here), the actual temperature into the melt close to the growth interface is lower than the liquidus temperature due to the presence in the melt of dopants or, in the case of a compound crystal growth, of a large excess of one component. The latter is the case of GaAs. A supercooled region
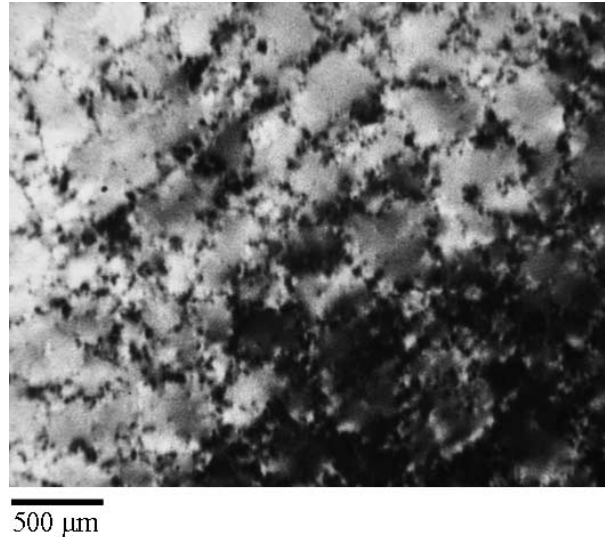
500 μm

Fig. 2. – 620 double-crystal topographs of a GaAs crystal to evidence the crystallites.

occurs when the thermal gradient at the interface G_L is lower than a value given by

$$(2) \qquad G_L \leq -v\frac{m_L}{D_L}\frac{1-k}{k}C_L,$$

where v is the growth rate, m_L is the slope of the liquidus temperature curve, k is the dopant distribution coefficient, and C_L is the dopant concentration in the melt. When this condition is satisfied, a condition of non-equilibrium occurs with the introduction of dislocations and formation of a cell structure, with every cell surrounded by a dislocation network. A more accurate analysis [21] showed that the surface energy has the effect to stabilize the interface even at low thermal gradient, so that the stability region is usually larger than forecasted by eq. (2).

In any case, eq. (2) suggests the growth parameters that control the formation of the cell structure. For example, a larger dopant concentration (or stoichiometry deviation), or an increase of the growth rate, or the choice of a dopant with large m_L and low distribution coefficient increase the cell formation.

However, another effect that can modify the cell structure should be taken into account. It is known that in the case of crystal growth by LEC method the thermo-mechanical stress reaches maximum values at the crystal surface and in the center [22]. Thus, due to the fact that dislocations are formed where the thermal stress overcomes a threshold value corresponding to the critical resolved shear stress, the dislocation density is expected to peak at the crystal borders and in the center, showing a typical W shape. Moreover, an increase of the stress together with the increase of the crystal diameter is also forecasted.
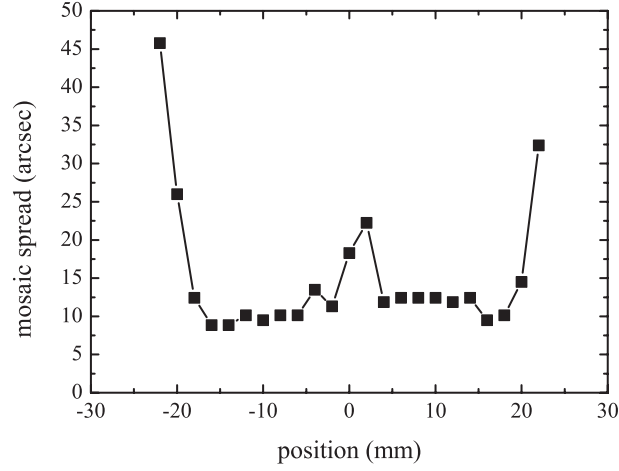
Fig. 3. – Mosaic spread of the GaAs sample 88-47.

4. – Results and discussion

4˙1. *Mosaic GaAs sample characterization*. – The mosaic spread of a crystal can be derived from high-resolution X-ray diffraction rocking curve measurements. The 004 Bragg diffraction profiles have been measured by setting the 2Θ position to the GaAs 004 peak and scanning the angle of the incidence ω near the maximum position.

We measured a FWHM of about 14 arcsec for a perfect GaAs Si-doped crystal, resulting from the convolution of the intrinsic 004 GaAs profile with the four crystal germanium 220 monochromator diffraction profile. In order to determine the mosaic spread, we assume that both this convoluted profile and the mosaic spread are described by Gaussian functions. Therefore, σ can be obtained from the experimental $\text{FWHM}_{\text{exper}}$ of the Bragg diffraction peak as

$$(3) \qquad\qquad \sigma = \sqrt{FWHM_{\text{exper}}^2 - FWHM_{\text{instr}}^2},$$

where we can assume $\text{FWHM}_{\text{instr}} = 14$ arcsec.

Figure 3 shows the mosaic spread of the undoped GaAs sample 88-47 as derived in accordance with eq. (3) from the rocking curves measured at different positions (a 2 mm sampling along a 44 mm range) along a wafer diameter. This mosaic spread follows the expected typical W shape often observed in undoped Czochralski grown GaAs crystals (see sect. **3**). According to previous studies [23], the σ in the sample central zone can be considered nearly uniform, with a mean value of 11 arcsec.

In fig. 4 the mean mosaic spread values as obtained from rocking curves measured on GaAs samples grown in different conditions (see sect. **2**) are reported. It is clear that both the growth conditions and the doping influence the mosaicity degree of the sample. The last point corresponds to sample Si9822-37, which is a wafer of GaAs doped with Si considered for comparison; as expected, its mosaicity is really low. This means that Si-doped GaAs samples are not suitable as optical elements in a Laue lens. Almost all the analyzed samples, not grown for this particular purpose, have a mosaic spread
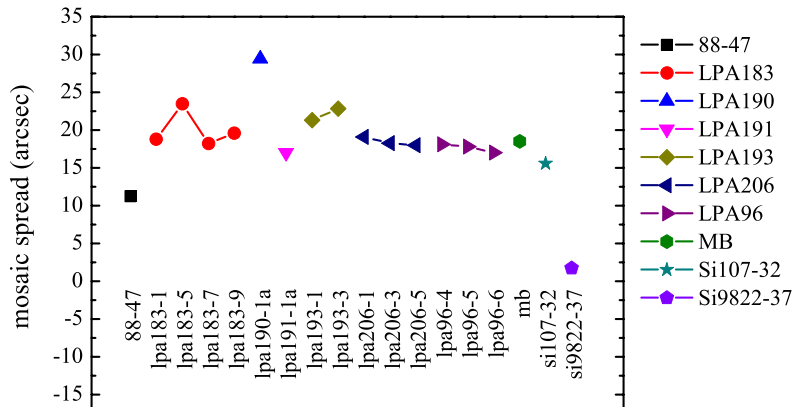
Fig. 4. – Mosaic spread of several GaAs samples grown in different conditions. The increasing suffix number corresponds to wafers cut at different heights, from the neck to the end, of the same ingot.

ranging from 10 to 25 arcsec, corresponding to the low limit end required for a Laue lens for hard X-rays with focussing distance of several tens meters; but in principle, it should be possible to obtain the requested characteristics by properly choosing the growth parameters in terms of thermal gradient and doping concentration.

4˙2. *Artificial mosaic GaAs samples*. – An alternative strategy to obtain crystals diffracting in a large angular range at high X-ray energy is to use curved crystals [1, 2, 7, 10, 11]. For curved crystals the diffraction range is given by the total curvature of the crystal lattice planes crossed by the X-ray beam. Provided that the lattice curvature radius R is larger than the ratio between the extinction length Λ and the intrinsic Darwin width of the Laue diffraction profile [16], the diffraction efficiency can reach values close to 100%.

Several GaAs crystals were afterwards treated to obtain curved crystals, that we call "artificial mosaic" crystals. A stack of these crystals, assembled as shown in fig. 5a, can be used as alternative optical element in a Laue lens. The lattice curvature can be induced by a surface damaging, which introduces defects in a superficial layer and creates an expansion of the crystal lattice. The curvature should then be spherical and the treated side should result convex. If t is the crystal length, the diffraction range α is given by t/R; if we consider, for example, a stack with $t = 1$ cm and $\alpha = 30$ arcsec, the desired radius is $R = 68.75$ m.

In the present case, the damaging was achieved by means of lapping with sandpaper on one side of a planar sample.

Also in this case high-resolution X-ray diffraction measurements allow to determine the local curvature radius of the sample by measuring the rocking curve angular shift as a function of the incidence position on the sample. Figure 5b shows the dependence of the local curvature radius R on the position on the sample surface of a GaAs sample $500 \, \mu$m thick before (black squares) and after (red circles) a treatment of only 5 minutes with sandpaper p1200. The mean radius changes from a value of about 100 m to 8 m. A longer treatment carried out on another GaAs sample $500 \, \mu$m thick causes an even bigger bending, with a final mean radius of about 2.8 m. Thicker samples can be treated
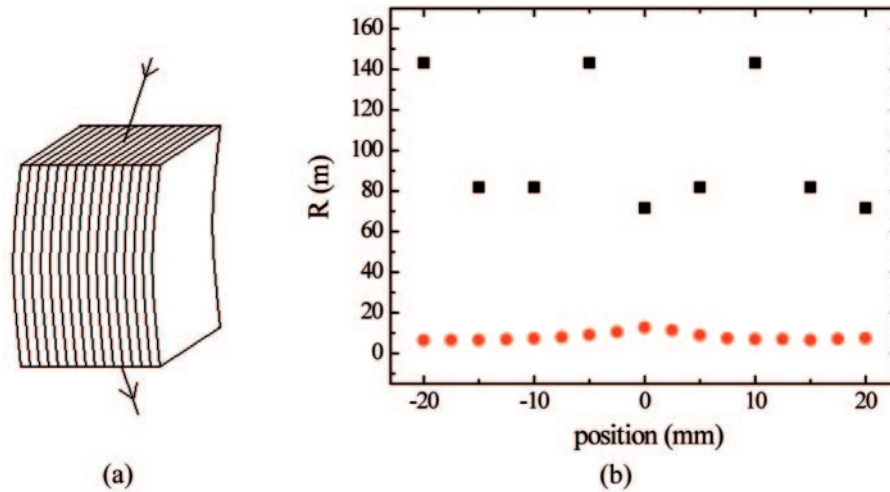
Fig. 5. – (Colour on-line) (a) Stack of curved crystals. (b) Curvature radii of the GaAs sample Si107-32 (sample $500\,\mu$m thick) as measured before (black squares) and after (red circles) a treatment of 5 minutes with sandpaper p1200.

with sandpaper of different grits and for various time lengths to achieve the desired curvature.

5. – Conclusions

In order to increase the collection efficiency of a lens for hard X-ray energies, mosaic crystals to be used as optical elements in Laue diffraction are proposed with mosaicity of the order of a few tens of arcseconds (a suitable mosaic angular spread depends on the desired energy resolution of the lens). Gallium arsenide crystals showed to be good candidates as optical elements. In fact, at variance with germanium, which has already been used for this purpose, GaAs shows a natural degree of mosaicity due to the spontaneous formation during the growth process of cellular structures with dislocations at the boundaries. Several GaAs samples grown by LEC method have been characterized by means of high-resolution X-ray diffraction. The mosaic spread values derived are slightly lower with respect to the requested one, but these crystals were not grown for this specific aim. Since proper growth conditions allow to control and modify both the dislocation density and the cellular structure responsible of the mosaic spread, the possibility of obtaining crystals with a given degree of mosaicity by tuning the LEC growth conditions is proposed.

We proposed also a complementary strategy to increase the Darwin width of the diffraction curve: the use of curved crystals as "artificial mosaic". More in details, we proposed an optical element constituted by a stack of curved crystals. The lattice curvature was achieved by introducing a compressive stress on the crystal surface by damaging it with fine sandpaper. Curvature radii between 2 and 8 m were easily obtained in wafer crystals $500\,\mu$m thick. We expect that a treatment of a thicker sample leads to higher radii.

REFERENCES

[1] BARRIÈRE N., VON BALLMOOS P., HALLOIN H., ABROSIMOV N., ALVAREZ J. M., ANDERSEN K., BASTIE P., BOGGS S., COURTOIS P., COURVOISIER T., HARRIS M., HERNANZ M., ISERN J., JEAN P., KNÖDLSEDER J., SKINNER G., SMITHER B., UBERTINI P., VEDRENNE G., WEIDENSPOINTNER G. and WUNDERER C., *Exp. Astron.*, **20** (2005) 269.

[2] SMITHER R. K., ABU SALEEM K., ROA D. E., BENO M. A., VON BALLMOOS P. and SKINNER G. K., *Exp. Astron.*, **20** (2005) 201.

[3] FRONTERA F., PISA A., LOFFREDO G., PELLICCIOTTA D., CARASSITI V., EVANGELISTI F., ANDERSEN K., COURTOIS P., AMATI L., CAROLI E., FRANCESCHINI T., LANDINI G., SILVESTRI S. and STEPHEN J., *Exp. Astron.*, **20** (2005) 241.

[4] HALLOIN H., *Exp. Astron.*, **20** (2005) 171.

[5] HALLOIN H. and BASTIE P., *Exp. Astron.*, **20** (2005) 151.

[6] VON BALLMOOS P., HALLOIN H., EVRARD J., SKINNER G., ABROSIMOV N., ALVAREZ J., BASTIE P., HAMELIN B., HERNANZ M., JEAN P., KNÖDLSEDER J., LONJOU V., SMITHER B. and VEDRENNE G., *New Astron. Rev.*, **48** (2004) 243.

[7] BARRIÈRE N., ROUSSELLE J., VON BALLMOOS P., ABROSIMOV N. V., COURTOIS P., BASTIE P., CAMUS T., JENTSCHEL M., KURLOV V. N., NATALUCCI L., ROUDIL G., FRISCH BREJNHOLT N. and SERRE D., *J. Appl. Crystallogr.*, **42** (2009) 834.

[8] PELLICCIOTTA D., FRONTERA F., LOFFREDO G., PISA A., ANDERSEN K., COURTOIS P., HAMELIN B., CARASSITI V., MELCHIORRI M. and SQUERZANTI S., *IEEE Trans. Nucl. Sci.*, **53** (2006) 253.

[9] ZACHARIASEN W. H., *Theory of X-Ray Diffraction in Crystals* (Dover Pubblications Inc., New York) 1945.

[10] SMITHER R., ABU SALEEM K., BENO M., KURTZ C. and KHOUNSARY A., *Rev. Sci. Instrum.*, **76** (2005) 123107.

[11] BARRIÈRE N., GUIDI V., BELLUCCI V., CAMATTARI R., BUSLAPS T., ROUSSELLE J., ROUDIL G., ARNAUD F. -X., BASTIE P. and NATALUCCI L., *J. Appl. Crystallogr.*, **43** (2010) 1519.

[12] IVANOV B. G. and KAEVA E. S., *Crystallogr. Rep.*, **50** (2005) S22.

[13] KOHNLE A., SMITHER R., GRABER T., FERNANDEZ P. and VON BALLMOOS P., *Nucl. Instrum. Methods Phys. Res., Sect. A*, **416** (1998) 493.

[14] COURTOIS P., BIGAULT T., ANDERSEN K. H., BAUDIN-CAVALLO J., BEN SAÏDANE K., BERNERON M., EL-AAZZOUZZI A., GORNY D., GRAF W., GUIBLAIN T., HEHN R., HETZLER E., MENTHONNEX C., MESTRALLET B. and DEWHURST C., *Phys. B*, **385–386** (2006) 1271.

[15] NYILAS K., DUPAS C., KRUML T., ZSOLDOS L., UNGÁR T. and MARTIN J. L., *Mater. Sci. Eng. A*, **387–389** (2004) 25.

[16] MALGRANGE C., *Cryst. Res. Technol.*, **37** (2002) 654.

[17] ZANOTTI L., in *Crystal growth in Science and Technology*, edited by AREND H. and HULLIGER J., *NATO ASI Ser. B: Physics*, Vol. **210** (Plenum Publishing Corporation, New York) 1990, pp. 347-357.

[18] DARWIN C. G., *Philos. Mag.*, **27** (1914) 315.

[19] HULL D. and BACON D., *Introduction to Dislocations* (Butterworth-Heinemann, Oxford) 2001.

[20] RUTTER J. E. and CHALMERS B., *Can. J. Phys.*, **33** (1953) 723.

[21] MULLINS W. W. and SEKERKA R. F., *J. Appl. Phys.*, **35** (1964) 444.

[22] JORDAN A. S., CARUSO R., VONNEIDA A. R. and NIELSEN J. W., *J. Appl. Phys.*, **52** (1981) 3331.

[23] FERRARI C., ZANOTTI L., ZAPPETTINI A. and ARUMAINATHAN S., *Proc. SPIE*, **7077** (2008) 70770O.