A CCD area detector for X-ray diffraction under high pressure for rotating anode source[†]

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Abstract. Details of a two-dimensional X-ray area detector developed using a charge coupled device, a image intensifier and a fibre optic taper are given. The detector system is especially optimized for angle dispersive X-ray diffraction set up using rotating anode generator as X-ray source. The performance of this detector was tested by successfully carrying out powder X-ray diffraction measurements on various materials such as intermetallics AuIn₂, AuGa₂, high Z material Pd and low Z scatterer adamantane (C₁₀H₁₆) at ambient conditions. Its utility for quick detection of phase transitions at high pressures with diamond anvil cell is demonstrated by reproducing the known pressure induced structural transitions in RbI, KI and a new structural phase transition in AuGa₂ above 10 GPa. Various softwares have also been developed to analyze data from this detector.

Keywords. CCD X-ray detector; X-ray diffraction; high pressure studies.

1. Introduction

The high sensitivity, wide dynamic range and high spatial resolution of image plate (IP) area detector are currently being exploited for investigation of materials under pressure to study phase transitions and equation of state. However, because of the small sample size in the high pressure diamond anvil cell (DAC), the recording time of the diffraction patterns with IP, is still a few hours with rotating anode X-ray source. The reduction in acquisition time is thus an important task. For this, various laboratories in the world are developing the charge coupled device (CCD) (Deacan *et al* 1990; Gago *et al* 1993; Hamaya *et al* 1996) based area detectors. These detectors show all the promising features of imaging plates combined with quick data collection ability. The availability of intense X-rays from synchrotron sources has opened up possibilities of real time investigations of materials in DAC using X-ray diffraction with CCD as area detector. However, the high cost of these detectors have limited their utilization (Fuchs *et al* 1990).

We have designed a low cost prototype CCD based area detector suited for rotating anode generator for fast data collection at normal and high pressure conditions for angle dispersive powder diffraction (ADXRD) studies and to provide definite directions in which the more accurate experiments employing IP can be carried out.

The performance of this detector system is established by carrying out angle dispersive X-ray diffraction measurements

on various substances like high atomic number element Pd, intermetallics like AuIn₂, AuGa₂ and low scatterer like molecular solid adamantane C₁₀H₁₆ at ambient conditions. Its utility for high pressure X-ray diffraction studies has been tested by reproducing the known pressure induced structural phase transitions in RbI and KI (Vaidya and Kennedy 1971). We have also used it for quick check of an unknown structural phase transition in the intermetallic compound AuGa₂.

2. Description of detector

The schematic diagram of the detector is shown in figure 1. The main parameters guiding the selection of components for designing such a CCD based detector system have been optical collection efficiency, optical demagnification, resolution, detection efficiency of the scintillating screen and its thickness. The detector uses a X-ray sensitive scintillator pasted on a taper fibre optic having input area of 70 mm coupled to a two-stage image intensifier tube. The output of image intensifier tube is transferred through a lens to a CCD imaging camera. The overall gain of the image intensifiers system which consists of two lens coupled image intensifiers can be manually adjusted depending on X-ray intensity. The two second generation image intensifier tubes have input/output diameter of 18 mm with 30 lp/mm spatial resolution (Sinha *et al* 1996). The CCD employed by us is 2/3 inch in diameter having 756 (H) × 581 (V) pixels with pixel size of 11.0 µm (H) × 11.0 µm (V). The video signals obtained from CCD are digitized using a 8 bit frame grabber with a 16 bit processor. We have introduced provision for online thresholding and online dark current subtraction. Shading

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correction to correct for the systematic nonuniformity of detector response was also incorporated. The image acquisition system can display real time images on a separate video monitor (40 ms) or display background subtracted images or integrated images using 16 bit onboard processor. We have also developed two custom designed algorithms to analyze the X-ray diffraction patterns. The first one iteratively determines the pixel position of the centre of the diffraction rings. The second algorithm carries out radial integration along the Debye–Sherrer rings.

3. Experimental

In order to study the performance of CCD area detector we have used it in place of IP detector in our ADXRD system (Godwal *et al* 1997). We used rotating anode X-ray generator operating at 50 KV and 40 mA to generate the X-rays. Zirconium filtered X-rays were collimated to 300 μm size at the samples. The overall resolution (pixel size) of the detector was 170 μm . In order to turn the radially integrated digitized image into a standard one-dimensional (1-D) powder pattern expressed in terms of 2θ , the sample to detector distance was accurately determined (~ 10 cm) by collecting the diffraction pattern of Pd at ambient conditions and from the knowledge of its lattice parameter (Pearson 1967).

4. Results and discussion

The typical patterns are shown for Pd and AuGa₂ in figures 2a–b. The strong powder diffraction rings from good scatterers like Pd and the intermetallics such as AuIn₂, and AuGa₂ were in fact observed at real time rate of 40 ms. The texture is seen in diffraction rings of Pd. Radially integrated diffraction patterns after background subtraction were transformed to one-dimensional pattern using the procedure described in § 3. In figures 3a–b, we display these diffraction patterns for AuGa₂ and KI. The data was fitted to yield the ambient values of the lattice

Figure 1. Schematic diagram of CCD based area detector for angle dispersive X-ray diffraction (ADXRD) experiments.

parameters of AuGa₂ and KI, respectively. An angular range of 2θ equal to 40° has been covered and if required, a step scan can be used to increase this to a higher desired value. A Rietveld refinement of all powder data with pseudo Voigt line profile yielded good agreement between the observed and simulated patterns (Rietveld 1967, 1969). In figure 4 we illustrate it for AuGa₂. The refined value of the lattice parameter for AuGa₂ was found to be

Figure 2. Online Debye–Sherrer rings for (a) palladium and (b) AuGa₂.

6.050 Å which can be compared with the published value (Pearson 1967) of 6.07 Å.

For high pressure measurements, RbI, KI and AuGa₂ samples were loaded in a modified Mao Bell diamond anvil cell (DAC) along with a ruby chip (Chidambaram

1992). Pressure was measured by ruby fluorescence technique (Mao *et al* 1996). The diffracted X-rays at various pressures were recorded on the CCD detector placed behind the DAC. In view of the limited opening of the DAC, the angular range (40°) of the detector does not

Figure 3. Radially integrated and background subtracted 1-D patterns for (a) AuGa₂ and (b) KI. The space groups and refined values of lattice parameters are also shown.

Figure 4. Rietveld refined pattern for AuGa₂.

Figure 5. (a) 1-D high pressure (3 GPa) X-ray diffraction pattern for KI. The indexing of various lines as seen in the figure is consistent with CsCl structure. (b) X-ray diffraction pattern for KI at ambient pressure in NaCl phase.

limit its utility for high pressure measurements. In figure 5 we show the one-dimensional diffraction patterns of KI when the pressure was increased to about 3.0 GPa. It is evident from the figure that KI has transformed to the CsCl structure from its ambient NaCl structure (Vaidya and Kennedy 1971). The measured value of the lattice parameter of 4.066 Å at 3 GPa pressure is consistent with the known experimental data (Vaidya and Kennedy 1971). On release of pressure KI returned to its ambient NaCl structure.

We have also used this system to look for quick signatures of the structural phase transition in the intermetallic compound AuGa₂ to the pressures of about 10 GPa (Alka et al 1998). The present high pressure X-ray diffraction measurements carried out using the CCD based X-ray imaging reveal the splitting of [200] line of the CaF₂ phase above 10 GPa similar to that observed by us in AuIn₂ (Godwal et al 1997). The subsequent detailed measurements carried out using IP has indeed confirmed this.

5. Conclusions

The performance of a CCD based area detector system developed by us is demonstrated at ambient and high pressure conditions using low current rotating anode X-ray source. The feasibility of reducing the data acquisition times of few hours (for e.g. while using IP area detector) to few tens of seconds is clearly established by collecting diffraction data, which can be Rietveld, refined. Considering the variety of samples and experiments for which this detector has been tested, it can be inferred that such detectors can play a vital role in rapid scan of various phase transitions in materials and provide the definite directions in which the more accurate experiments employing IP can be carried out. This detector may also be suited for certain new class of experiments such as time resolved phase transitions which are not feasible with the present form of an IP area detector. If the current CCD is replaced by a cooled one then higher dynamic range and precision can be obtained.

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