Conf-8809197

TWO DIMENSIONAL CCD ARRAYS AS PARALLEL DETECTORS IN ELECTRON ENERGY LOSS AND X-RAY WAVELENGTH DISPERSIVE SPECTROSCOPY*

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CONF-8802127--2

DE89 003653

AUGUST 1988

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*Work supported by the U.S. Department of Energy, BES-Materials Sciences, under Contract W-31-109-Eng-38.

This manuscript was presented at the Workshop on "Electron-Beam Induced Spectroscopies at Very High Spatial Resolution", Aussois, France, March 1988.

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Two Dimensional CCD Arrays as Parallel Detectors in Electron Energy Loss and X-Ray Wavelength Dispersive Spectroscopy

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Abstract

Parallel detection systems for spectroscopy have generally based upon linear detector arrays. Replacing the linear arrays with two dimensional systems yields more complicated devices however there are corresponding benefits which can be realized for both x-ray and electron energy loss spectroscopy. The operational design of these systems as well as preliminary results from the construction of such a device used for electron spectroscopy are presented.

Introduction

The merits of using parallel detection scheme for electron spectroscopy have been reviewed by a variety of authors [1-5] as well as in these proceedings. In this paper we will consider the specific topic of replacing conventional linear photodiode (PD) and charged coupled device (CCD) linear arrays with a two dimensional (2-D) CCD array. 2-D CCD's of research quality can be procured in array sizes ranging from 128^2 to 2048^2 pixels. In this work we shall be considering designs based upon a high sensitivity, high resolution CCD, which is used in medical or astronomic imaging namely a Tektronics TK512M 512^2 pixel CCD array and is shown in figure 1. The sensitive area of the array is 13.8 x 13.8 mm^2 and has square pixel elements which are approximately 27 μm in width. The CCD controller, detector head electronics and electron optics were developed at ANL specifically for high speed data acquisition and allow the recording of spectra in as short a time as 10 μsec or approximately 1000 times faster than the typical 1024 pixel photodiode arrays.

2-D CCD systems can be operated in two modes: as a pseudo-linear device for spectral acquisition or as a conventional two dimensional imaging system, both of which will be illustrated in this work. When operating in a spectral acquisition mode an adjustable mask covers most of the CCD array area, and a deflector system is used to shift the spectrum over a small window frame (~ 5 x 512 pixels) of the array which is used for data collection, as illustrated in figure 2. When a pixel well reaches saturation (~690K electrons , ~265K photons) the entire spectral frame is electronically shifted down the CCD array to the masked (i.e. unused) region of the array which essentially acts a massive data buffer. Spectra are digitized by an on-board 15 bit ADC and transferred to a DEC MicroVax II computer system using a DRV11 parallel interface board and software developed at ANL. Operation as an imaging detector requires that the mask be removed and that the image is blanked during the CCD readout operation.

All operating parameters of the CCD such as frame size, readout rate and exposure time are downloaded by the host computer into the CCD controller (figure 3). The maximum transfer rate in the horizontal shift register is 5 Mhz which for a 512^2 element detector gives ~ 50 msec transfer time. Although data transfer rates within the CCD are high (~1 Mhz down the array and 5 Mhz through the horizontal shift register) the limiting factor in a complete CCD data transfer becomes the ADC digitization time constant, which varies depending upon the digitization depth (i.e 12, 14, 16 ... bits). For 15 bit resolution time constants on the order of 10 μsec are attainable. At this digitization rate the full spectral dump of 100 frames x 512 pixels requires ~0.5 sec (we choose to bin spectra in ~ 5 x 512 wide frames). Alternatively the

CCD can be software programed to perform a nearly continuous but slower transfer of its contents providing the illusion of real time spectral acquisition.

This data transfer and digitization process is a real limitation in the usage of any type of array device when employed as "parallel detection" system. A true parallel device requires a single video amplifier per pixel column and has only been achieved for arrays of size up to 64^2 which is insufficient resolution for AEM based applications. In the current generation of array detector, regardless of whether it is a linear or a 2-D array, the measured intensity in the device must be serially dumped through a horizontal shift register, through a single video amplifier and then into a single ADC. Thus, in simple linear PD or CCD devices, one cannot acquire time resolved spectra faster than the ADC digitization rate or about 10 msec for a 1024 element linear array. For time resolved studies this becomes the rate limiting step. In contrast, using a 2-D device having a masked zone as a data buffer allows time resolutions of ~10 μsec to be attainable over the temporal window required to fill the entire array with data. Clearly, the data transfer rate to the display/analysis computer system will be slower owing to the greater amount of information which is present in the device, however, it will no worse than than of the linear array, scaled by the increased pixel number density. Should even faster time resolutions be required for a particularly demanding application, then the mask can be removed from the entire 2-D array and a deflection system (figure 4) used to convert the CCD array into the equivalent of a streak camera. The time resolution now being limited only by the deflection of the spectra across the CCD array face.

Application to Electron Energy Loss Spectroscopy

For use in electron spectroscopy, a 2-D CCD system is configured so that it views a loss spectrum in nearly the same way as a conventional linear photodiode array [3,7]. Spectra are magnified by quadrupole lens system attached to a magnetic sector electron spectrometer (Gatan 607) beneath an AEM (Philips EM420) as is illustrated in figure 4. The spectra impinge upon a Ce:YAG scintillator and are then transferred by a unit magnification f/1.2 optical lens system to the CCD array, which is thermoelectrically cooled to -30^{o} C to reduce noise. (This lens system is presently being replaced by a fiber optics bundle.) A conventional electromagnetic deflection system is used to align the spectra with the spectral window frame used for digitization as discussed previously. Figure 5 shows an example of a digitized B K edge from Boron Nitride energy loss spectrum. The data is shown both as an image of the frame shifted spectra accumulated in the CCD device as well as a line profile through a single CCD pixel row. For sake of clarity only a small portion of the CCD image is shown (~70 x 100 pixels). Using a simple software program, successive spectra can be summed to produce a line profile spectrum as would be obtained using a serial detector.

Another advantage which is derived through the use of a 2-D array over that of a linear photodiode is the fact that by removing the CCD mask the entire device becomes an active imaging detector system. This for example, allows for spectrometer alignment by the dynamic viewing of the spectrometer aberration figures. In addition, by the appropriate electron-optical lens adjustment, the CCD can be utilized as an imaging device. Figure 6 illustrates the acquisition of an unfiltered [001] electron diffraction from a TEM specimen of Aluminium. The weak unidirectional streaks extending outward from each Bragg reflection are images of the EEL spectrum at each reflection. By the judicious introduction of an aperture in a conjugate image plane before the quadrupole lens system, energy filtered diffraction patterns (or images) can be obtained.

The Tektronics CCD system described above is a second generation device and is currently being tested on the AEM, hence its absolute performance relative to that of the prototype RCA SID 501EX CCD previously employed [7] has not yet been completely determined. It is expected that the Tektronics CCD will exceed that of the earlier RCA device, which had the following characteristics measured for 100 kV electrons at a beam current of 0.5 nA:

Conversion Efficiency: CCD electrons/ Incident electron	2.6
YAG:Ce Scintillation efficiency (photons/electron)	500
Statistical uncertainty of Output vs. Input	1.5
Detective Quantum Efficiency	0.4
Frame - to -Frame Pixel Fluctuation ($\sigma\%$)	0.4
Dynamic Range: Saturation peak/RMS noise	7000
RMS Dark Current	40 e^-
RMS Readout Noise	50 e^-
RMS Digitization Noise	45 e^-

Here, the conversion efficiency of the CCD system was measured by monitoring the incident beam intensity using a Faraday cup in the electron microscope, and measuring the integrated number of CCD output electrons from the detector. Using these values, and the formulae given by Deckman and Gruner [8] we calculated an experimental value of 0.4 for the detective quantum efficiency (DQE) of the system. The dynamic range of the detector was determined by measuring the ratio of the integral of the zero loss peak (in a 4x5 pixel window) to the square root of the noise in a 20 pixel background region and yields a dynamic range of ~7000. If M frames are summed then the dynamic range increases in proportion to $M^{1/2}$.

Application to X-ray Analysis in Wavelength Dispersive Spectroscopy

There has been renewed interest in wavelength dispersive spectrometers for light element x-ray analysis in the AEM, owing to their inherently higher energy resolution (~1 eV) in contrast to the poor resolution of solid state energy dispersive spectrometers (~70-100 eV). The conventional thought in this regard it to position miniaturized versions of curved crystal spectrometers as employed in electron microprobes into the column of a modern AEM [6]. These devices have two major disadvantages relative to their use in an AEM. First, they rely on precise movement of a detector and analyzing crystal system to scan a complete spectrum. Assuming that the device is not prone to mechanical failure, such motion may introduce serious vibration problems during the scanning process, and can therefore affect probe or stage positioning at the highest spatial resolution. Secondly, they are serial devices allowing only one x-ray line to be analyzed for each detector.

Let us now consider the problem from a new approach namely: that we wish to devise a parallel detection wavelength dispersive spectrometer (WDS) in the AEM with no moving parts. Referring to the diagram shown in figure 7, assume that we affix a simple flat analyzing crystal to the bottom of the upper pole piece of the objective lens of the TEM. In this geometry, x-rays leaving the specimen will be diffracted downward in conical ray paths each of which has a unique (λ,θ) relationship based upon Bragg's law. If one were to position a 2-D CCD detector system at the center of the lens gap, then one could intercept the Bragg diffracted x-rays and map out their angular distribution as shown in figure 8. At this location the detector would be shielded from direct electron damage, and still allow the insertion of a conventional Si(Li) EDS system in the optimum (downward) looking configuration as well as permitting all the normal stage motions and tilts. Since characteristic x-rays will be diffracted in conical sections the collection efficiency can be quite high (figure 8) eliminating the tedious process of curving the analyzing crystal to increase collection efficiency as is done in conventional WDS analyzers. Should higher collection efficiencies be required, then larger (1024^2) or multiple detectors can be located strategically around the specimen stage. Since there are no moving parts spatial resolution of the AEM will not be impeded due to vibration and the data collection will be in parallel. Computer programs can be written to map the pixel location to wavelength for quantitative analysis. This will be required as there will be a curvilinear data distribution on the CCD array face.

Inserting typical dimensions in the diagram of figure 7, for H ~ 7 mm and L ~ 4 mm, then the maximum Bragg angle one might achieve is θ_1~ 60°, while the minimum angle one might reach for a 13.8 mm long detector is θ_2~ 20°. Assuming an analyzing crystal having a d-spacing of ~2 nm (for example Mica, or a multilayered artificial crystal [9]), one calculates these Bragg angles as defining the wavelength range of 3.46 nm $\leq \lambda \leq$ 1.36 nm (360 eV $\leq$ E $\leq$ 910 keV) or approximately the elemental range of nitrogen to neon. Decreasing the analyzing crystal d-spacing will shift the detection range upwards in Z and vice versa. For example, if d is changed to 1.5 nm then the range of 26 nm $\leq \lambda \leq$ 1.02 nm (477 eV $\leq$E$\leq$ 1.21 keV) gives the range oxygen through sodium. To expand the wavelength range, three alternatives come to mind: 1.) the detector can be shifted horizontally, 2.) a larger CCD may be employed or 3.) the single analyzing crystal could be replace by two crystals (of different d-spacing) mounted side by side. In the first case one has limited utility, the second will both increase the collection solid angle as well as increase the spectral range but at a substantially higher cost (the cost per pixel for a CCD is about constant). The last method essentially reduces the collection solid angle by a factor of two, however, it in effect converts the CCD into a dual spectrometer system where on each of two halves of the CCD array a different wavelength regime is imaged by the two analyzing crystals. This may well prove to be an optimum configuration providing both high resolution, parallel detection and extended wavelength ranges in a single device. In all these scenerios, the wavelength (energy) resolution should be limited only by x-rays scattering from neighboring wells within the CCD which would cause excitation of the adjacent pixels. The inherent dispersion without this effect should be excellent (~1 eV/pixel at 500 eV).

Several practical difficulties will, of course, have to be addressed. One must re-engineer the CCD mount, control electronics, and thermoelectric cooler to fit within the allotted space in the pole piece gap, a difficult but not insurmountable problem. A beam blanking system will have to be part of the instrument to allow readout of the array without data overflow, and an appropriate scintillator developed for the CCD face plate which will have conversion efficiency near unity for the x-ray energies of interest. These problems in mind, the application of CCD technology still becomes an interesting and potentially powerful alternative to the conventional WDS curved crystal spectrometer systems for application to the next generation AEM.

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Figure Captions

Figure 1. Tektronics 512M - 512 x 512 CCD

Figure 2. CCD/Mask Configuration when device is used for spectral acquisition .

Figure 3. Schematic Diagram of Data Transfer in CCD and Controller

Figure 4. Electron Spectrometer/ CCD detector system used for EELS

Figure 5. Partial EEL spectrum from BN showing CCD images of the B K edge and a line profile through a single pixel row.

Figure 6. [001] Diffraction pattern of Al foil recorded through the EELS system, note the unidirectional streaks corresponding to EEL profiles on each Bragg reflection.

Figure 7. Schematic Diagram of a WDS system for X-ray Spectroscopy employing 2D-CCD arrays for parallel detection.

Figure 8. Projected distribution of diffracted x-rays on the CCD/WDS array face, illustrating the intensity distribution and collection solid angle. Incident electron beam is into the plane of the figure.

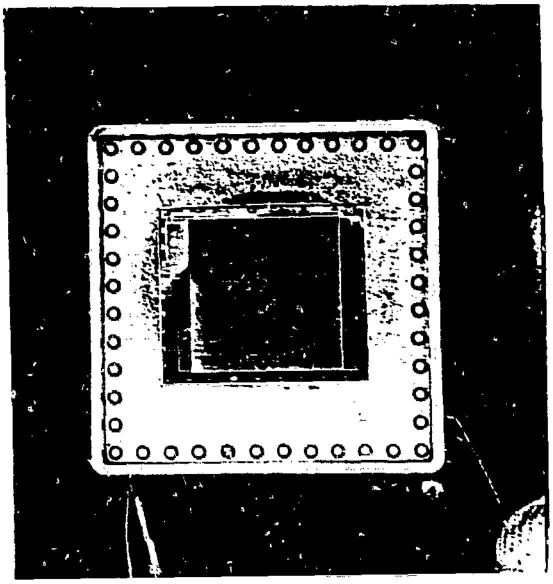

Figure 1 Tektronics 512M 512 x 512 CCD 13.8mm x 13.8mm, pixel size 27 microns

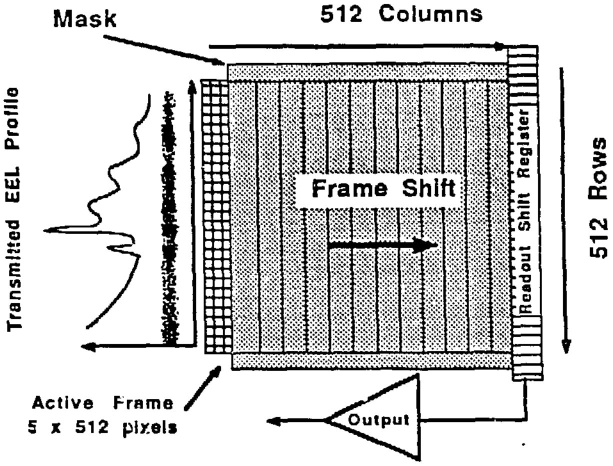

Figure 2. CCD Mask Configuration used during Spectral Acquisition.

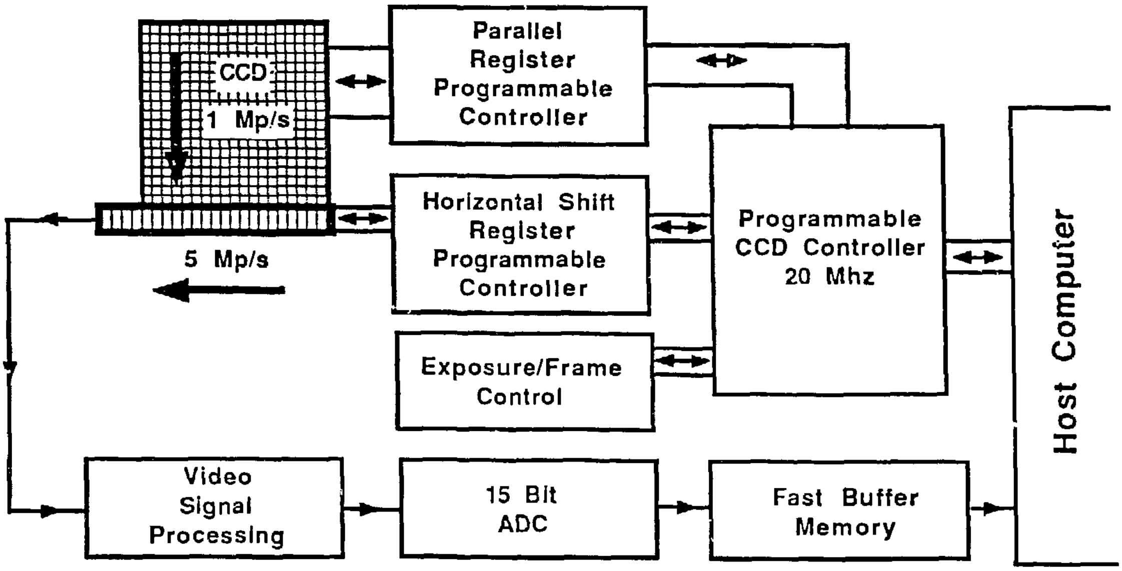

Figure 3. Block Diagram of CCD Controller and Data Paths

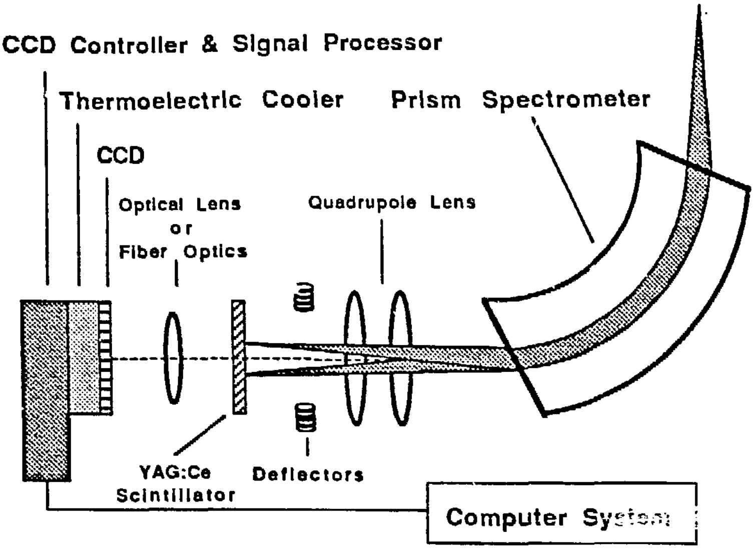

Figure 4. Electron Spectrometer
CCD Detector Configuration

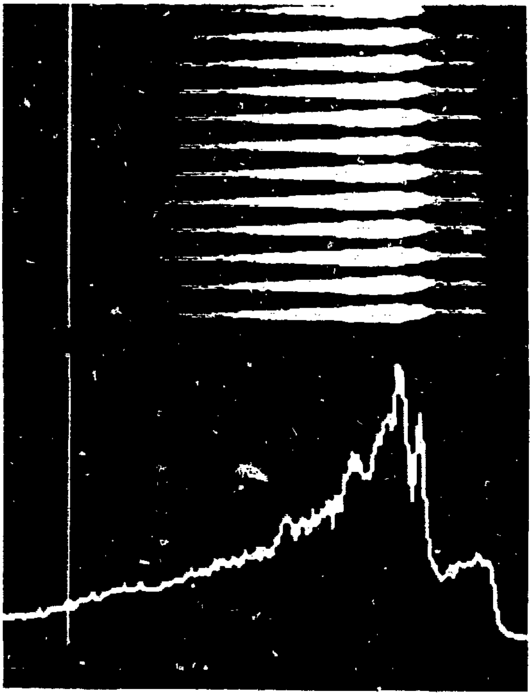

Figure 5. Partial EEL Spectrum of the B K edge in BN showing both intensity distribution within the CCD and a line scan through a pixel row.

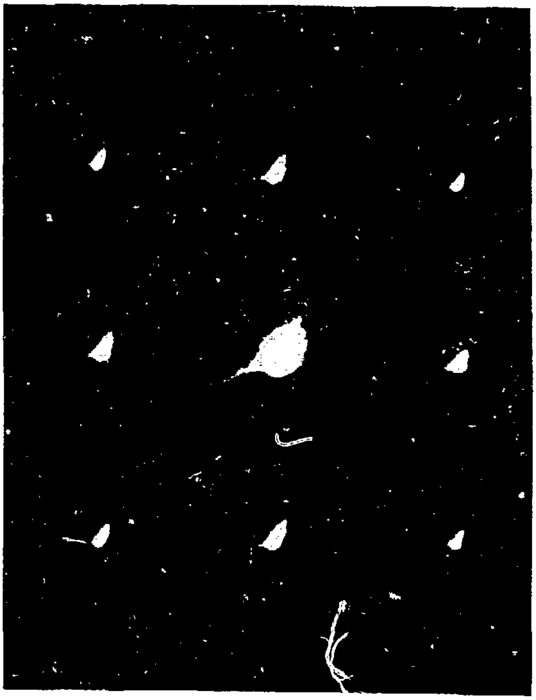

Figure 6. [001] Al diffraction pattern recorded through EELS/CCD system. Note unidirectional streaks corresponding to Loss Spectra at each Bragg Reflection.

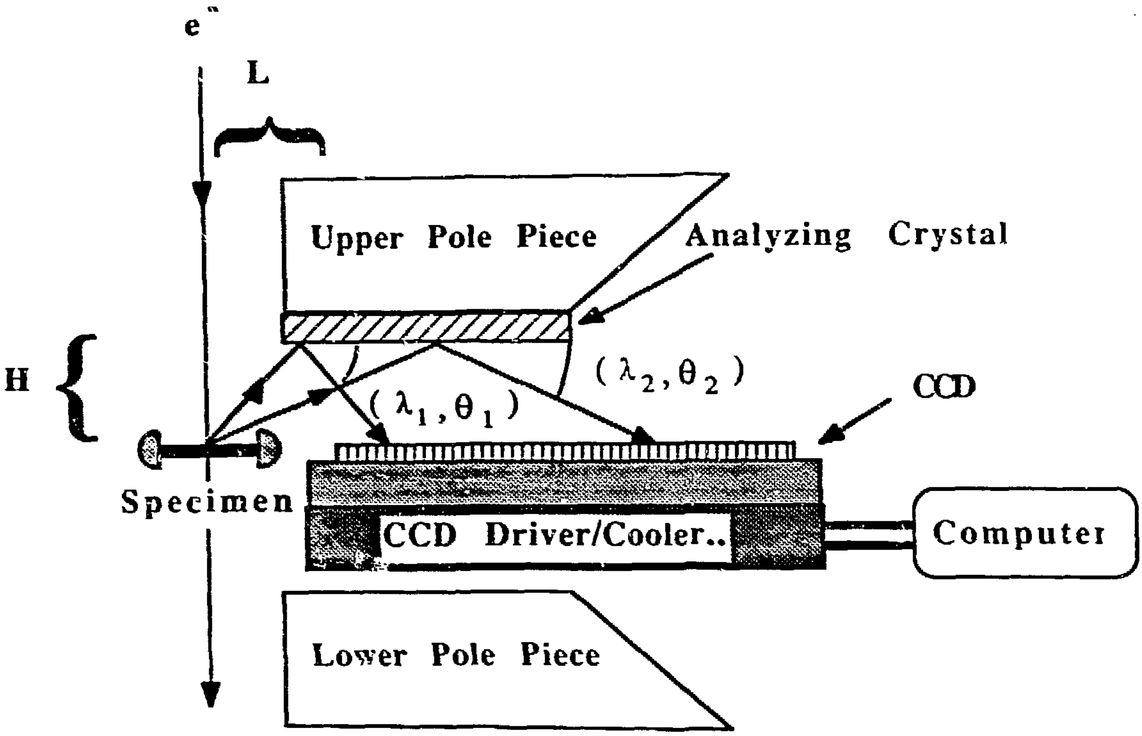

Figure 7. Schematic diagram of CCD/WDS detector system

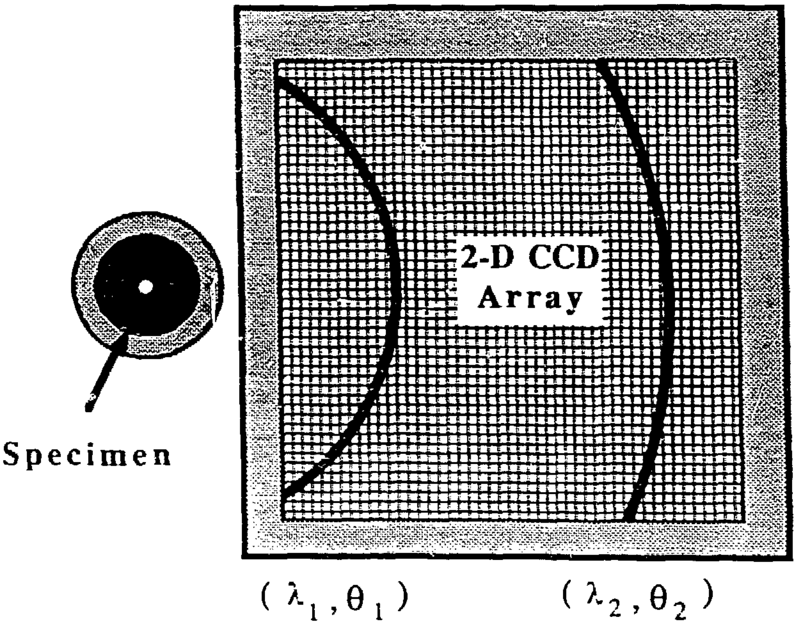

Figure 8. View of CCD/WDS system along the incident beam direction illustrating diffracted intensity distribution and the collection solid angle defined by the CCD array