

APPLICATION OF CCDs AS ELECTRON DETECTORS IN ELECTRON MICROSCOPY

P.T.E. Roberts, A.M. MacLeod, J.N. Chapman

ABSTRACT

A CCD area imaging array has been used as a detector of 20-100keV electrons to form the basis of an electronic imaging system for an electron microscope. This system has considerable advantages over conventional imaging methods for image processing applications and in the electron microscopy of radiation sensitive materials. This paper presents the results of preliminary experiments to measure CCD gain and the degradation in performance caused by irradiation. Electron images of specimens in the electron microscope have been obtained with 60keV electrons.

INTRODUCTION

In a conventional transmission electron microscope thin film specimens are illuminated uniformly with a monoenergetic electron beam and the resulting image is either viewed on a phosphor screen or recorded permanently on photographic emulsion. An electronic imaging system based on a CCD offers several advantages over the conventional system, particularly for image processing applications and when radiation sensitive specimens are under investigation. For the former case, the output of the CCD can be readily digitized to give an immediate quantitative record of the electron intensity distribution in the image in a form suitable for computer analysis. This is in marked contrast to photographic emulsion which requires considerable processing before quantitative data are available. For the latter application, where minimisation of the electron dose on radiation sensitive specimens is of the utmost importance, advantages accrue from the increased efficiency of electron detection of the CCD over a phosphor screen, together with the ability to store and continuously display images without continuing electron irradiation.

IMAGING SYSTEM

A block diagram of the imaging system is shown in fig. 1. The CCD is housed under vacuum in the camera chamber of a JEOL JEM 100C electron microscope which can be operated with electron energies from 20 to 100keV. The device used in these experiments is a Fairchild CCD 202, a 100x100 area imaging array, with an interline transfer structure. An electron image is incident directly on the front face of the device, and secondary electrons from the resultant electron-hole pairs produced in the silicon substrate are collected as charge packets in the irradiated cells. The charge collected in each cell is proportional to the electron flux incident on it. This charge is transported through the device to produce a series of voltage levels at the output, which are transferred to a dedicated Data Accumulation and Control Unit incorporating a Fairchild CCD Driver Board, with facilities for digitization, accumulation of data from

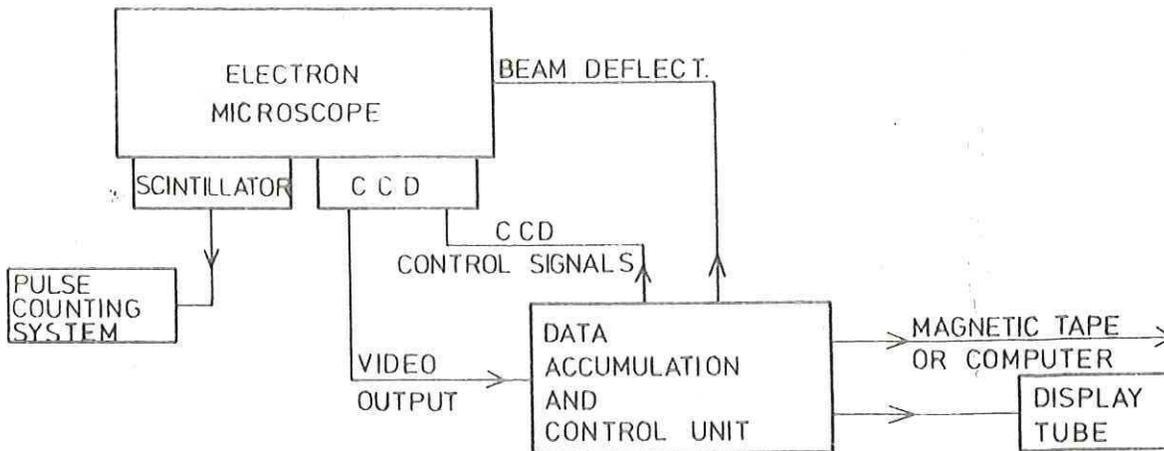


Figure 1: Block diagram of the imaging system

several identical images, subtraction of background and storage of the accumulated image in a solid state memory. The stored data can be used to form an image on a display tube and transferred to magnetic tape or a computer for further processing.

Substantial differences exist between using a CCD 202 for photon and electron imaging. For the latter, removal of the protective glass window is necessary to avoid absorption and scattering of the electron beam. Furthermore, the aluminium layer which shields the transport cells from light is virtually transparent to electrons in the energy range of interest, so, to avoid image smearing during transfer, the microscope is operated in a pulsed beam mode. Firstly, the Data Accumulation and Control Unit deflects the electron beam onto the specimen, simultaneously stopping the clocking signals to the CCD. After a preset time the electron beam is removed from the specimen, clocking is restored and, without affecting the charge stored in the photocells, the information in the transport cells is read out. With the transport cells cleared in this way, charge in the photocells is read out in the usual way for two interlaced frames. The sequence is repeated starting with the irradiation of the CCD by the electron beam, and the data from each cycle is superimposed in the Data Accumulation Unit to form an image. The system allows for storage either of a 100x100 array of photocells alone, or of an 82x82 array of photocells with the corresponding transport cells.

CCD PERFORMANCE DURING ELECTRON IRRADIATION

Before using the CCD for electron imaging, preliminary experiments were performed to measure the gain of the device and the effect of cumulative electron dose on its performance. Measurements were made at three electron energies, 20, 60 and 100keV. The number of electrons incident on the CCD was monitored by a scintillator-photomultiplier arrangement connected to a pulse counting system (fig.1). The scintillator was adjacent to the CCD and received the same electron flux.

GAIN The gain of the CCD was studied by flooding it and the scintillator with an electron beam of uniform intensity and measuring the average height of the output pulses from each cell of the CCD, a quantity

proportional to the average charge collected by a cell. The results of this experiment can be summarized as follows:

(i) For any electron energy the gain of the CCD (which we define as the number of secondary electrons collected for every incident primary electron) was independent of incident electron intensity, provided the cell content remained below its saturation value.

(ii) In the incident energy range from 20-100keV the gain increased non-linearly with energy, the gains for 20, 60 and 100keV electrons being 0.25×10^4 , 1.6×10^4 , 2.5×10^4 respectively. The number of secondary electrons collected for every incident primary electron is approximately proportional to the energy deposited within a diffusion length ($\approx 50\mu\text{m}$) of the semiconductor interface. Two factors act to reduce this below the incident electron energy and are responsible for the non-linearity referred to above. Firstly, energy is lost by the incident electron in passing through the overlying layers of the device. This increases as the incident electron energy decreases and is therefore greatest for 20keV electrons. Secondly, incident electrons of energy $>100\text{keV}$ have a range in silicon which is $>50\mu\text{m}$ and therefore produce electron-hole pairs which may not be collected at the interface. The gain of the CCD is therefore expected to decrease as the incident electron energy is increased substantially above 100keV.

CELL CAPACITY The number of incident electrons which cause cell saturation is low, since the number of secondary electrons each cell can contain is $\approx 10^6$ and the gain of the device as an electron detector is high. The cell capacities for 20, 60 and 100keV electrons correspond to 440, 65, and 45 incident electrons respectively.

As the number of incident electrons/cell should be ≥ 400 if random variations in the image are to be unimportant, each image involves several cycles of illumination, readout and accumulation. For example, using 60keV electrons, 8 cycles are needed to produce an acceptable image.

EFFECT OF CUMULATIVE DOSE By shielding part of the CCD from the incident electron beam it was possible to compare the performance of irradiated and unirradiated cells when uniformly illuminated with light from a LED. As the cumulative dose increased, two effects attributable to radiation damage were clearly observed.

(i) The dark voltage (i.e. the output voltage produced by the CCD when it is not illuminated) increased with cumulative dose, rising to 10% of the saturation voltage after 10^6 incident 60keV electrons/cell. By cooling the device to -40°C this voltage was substantially reduced so that its effect was negligible.

(ii) As cumulative dose increased, the gain of the CCD was no longer independent of incident electron intensity but decreased as the output voltage approached its saturation value. Restoration to its original level was achieved, however, by increasing the voltage applied to the gate electrode which increased the depth of the potential well. This was only possible for relatively low doses and, as the gate voltage could not be increased indefinitely, a decrease in gain which could not be simply corrected was ultimately observed. For each value of cumulative dose a maximum output voltage could be determined below which the gain was normal, and its variation with cumulative dose is shown as a function of incident electron energy in fig. 2. If the lifetime of the CCD is defined as the dose at which the maximum output voltage falls to half of

the original saturation output voltage, then the lifetime of the CCD was 2×10^6 , 1.2×10^7 and 1.5×10^7 incident electrons/cell for 20, 60 and 100keV electrons respectively. Since $\sim 10^3$ electrons/cell are desirable for most image analysis procedures in electron microscopy the lifetime of the CCD for 60keV electrons corresponds to 1.2×10^4 recorded images.

From these results it can be seen that the damage done to the CCD by an incident electron increases with decreasing energy. This is consistent with the suggestion that the degradation in CCD performance is due to radiation-induced positive charge in the SiO_2 layer, for the energy deposited in this layer increases as the incident electron energy falls from 100 to 20keV. This mechanism has been proposed by Killiany et al^{1,2} who noted similar degradation in the performance of a Fairchild 100x100 area imaging array under gamma irradiation. An improvement in CCD lifetime has been achieved by the use of a device with a more suitable structure using a radiation hard fabrication process^{3,4}. Furthermore it has been demonstrated that in MOS structures trapped positive charge has been removed by heating to temperatures $> 150^\circ\text{C}$ ⁵. If the CCD can be heated to this temperature without damage it may be possible to extend the lifetime of the devices by regularly heating after use to anneal out radiation damage.

ELECTRON IMAGES

Figs. 3 and 4 show electron images obtained using 60keV electrons with the CCD cooled to -40°C . To avoid damage to the on-chip amplifier, the outer area of the device was shielded from the electron beam by a copper mask, leaving an inner area of approximately 82×82 photocells and 82×42 transport cells to record the electron images. Fig. 3 shows the image of a $0.5 \mu\text{m}$ repeat replica diffraction grating, taken with an average number of electrons incident on each photocell of 630. To show that useful information is not restricted to the photocells fig. 3b shows the image formed from the content of the transport cells. Fig. 4 shows a through focus series of images of a perforated carbon film, each recorded with an average dose of 300 primary electrons/photocell. The Fresnel fringe on the overfocussed image is particularly prominent and its sharpness indicates that, at least for 60keV electrons the resolution of the image is limited principally by the size of the photocells ($\approx 400 \mu\text{m}^2$). As the electron energy is increased further, however, the diameter of the cloud of electron-hole pairs produced by electrons incident on one point will also increase⁶ and this effect is likely to limit the resolution. Finally it should be noted that whilst $\geq 10^3$ incident electrons/photocell are required to obtain good image intensity data for analysis, much smaller doses (< 50 electrons/photocell) may be sufficient to locate an area of interest and to adjust the focus. This allows radiation sensitive specimens to be imaged with very little preliminary irradiation during setting up.

CONCLUSIONS

Despite the fact that the CCD used in the experiments reported here was designed for photon imaging applications, its advantages as an electron detector in the conventional transmission electron microscope appear to be considerable. Its high gain when bombarded with electrons in the energy range of interest, together with its low noise properties,

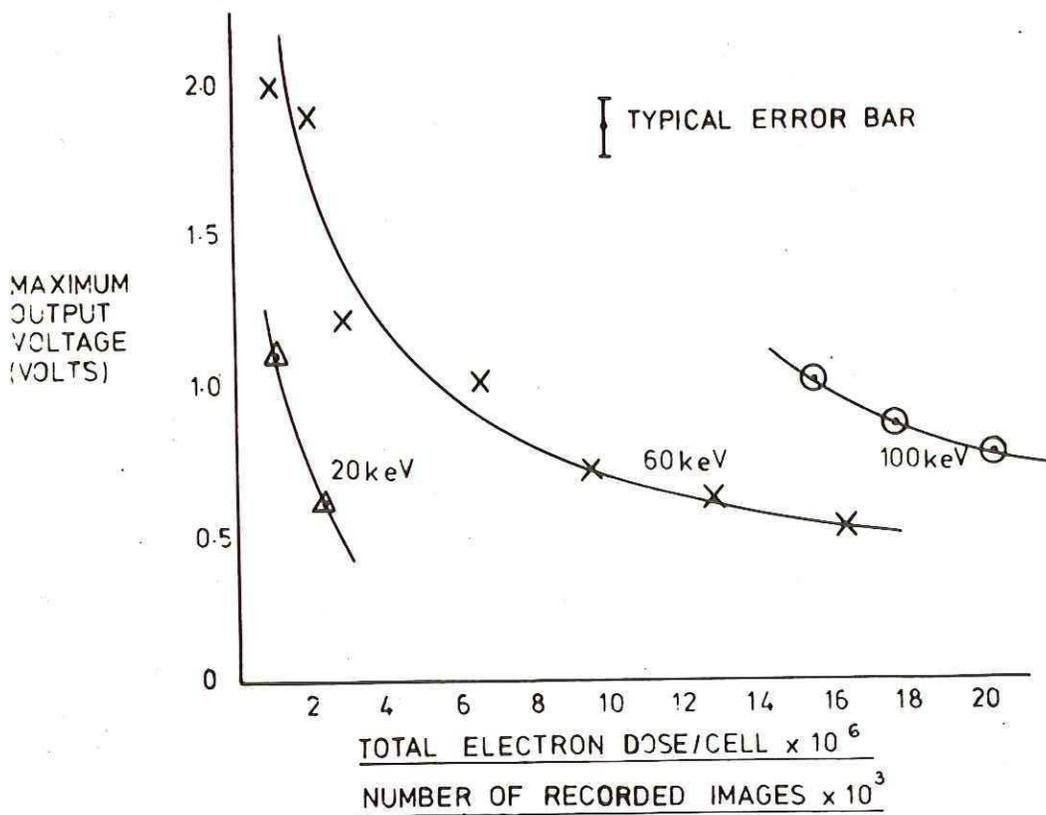
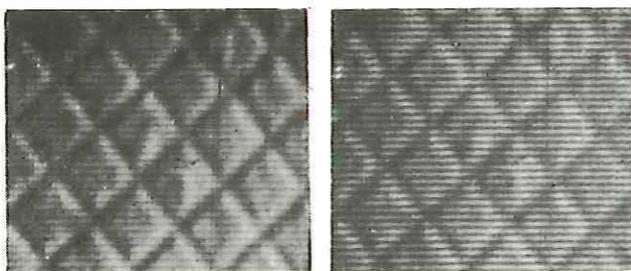


Figure 2: Variation in signal handling capability of CCD with cumulative electron dose for various incident electron energies. The gain of the CCD is decreased for signals greater than the maximum output voltage.



a) Photocells

b) Transport cells

Figure 3: Transmission electron image of a replica diffraction grating using 60keV electrons. Number of electrons/photocell = 630



a) Under focus

b) In focus

c) Over focus

Figure 4: Transmission electron image of perforated carbon film using 60 keV electrons showing Fresnel fringe. Number of electrons/photocell = 300

ensures that it acts as a detector with a high quantum efficiency so that recorded image intensities should be almost shot noise limited. Its most serious limitation when used to record electron images of the kind illustrated here is its susceptibility to radiation damage although a large number of images with sufficient statistical accuracy for most image analysis applications may be recorded in its lifetime. Nonetheless, to maximize the number of images produced with each device, approximate location of areas of interest and some focussing must be done using the relatively insensitive phosphor screen of the microscope. Thus the availability of radiation resistant CCDs, whilst advantageous for all applications in electron microscopy, would be of particular value when investigating radiation sensitive specimens where high efficiency electron detectors are desirable at all times.

Finally, it should be noted that the need for a parallel recording system with an electronic output is not restricted to image analysis in the conventional transmission electron microscope. Electrons with similar energies are used in scanning and scanning transmission electron microscopes and in electron diffractometers and spectrometers. It is possible that electronic recording systems based on CCDs will be beneficial in all these instruments and will greatly facilitate the collection of quantitative intensity data in a form suitable for immediate computer analysis.

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