Cathodoluminescence Measurements of CdTe in Transmission Electron Microscope

Wei-Chang D. Yang^{1,2}, Yohan Yoon^{1,2}, Benoit H. Gaury^{1,2}, Paul M. Haney¹, Nikolai Zhitenev¹ and Renu Sharma¹

Cathodoluminescence (CL) is an important spectroscopic method for characterizing photovoltaic materials in electron microscopes. When electron-generated free carriers recombine, CL signals are emitted from the luminescent material and provide spectroscopic information that can be used to reveal features of the electronic structure, such as the band gap and defect states near the band edge. These characteristics can be correlated with the microstructure and microstructural defects that limit the solar cell efficiency. Ultimately, this knowledge can be used to develop processing schemes that optimize material structure and performance. For example, grain boundaries create interfaces in the bulk of thin-film solar cells, leading to interfacial states may have an impact on the bulk recombination events and rates[1,2]. When investigating the influence of grain boundaries in scanning electron microscopes, the spatial resolution of CL is limited by the lateral size of the interaction volume (at least 250 nm). Much higher resolutions are needed to extract quantitative information from individual grain boundaries. Therefore, it is necessary to use high-spatial-resolution CL so that the change in electronic structure due to the interfacial states can be characterized.

In the present work, we use a home-built CL spectroscopy set-up integrated with a transmission electron microscope (TEM) to measure CL spectra of polycrystalline CdTe at room temperature with interaction volumes of ≈ 10 nm in diameter, as shown by Monte Carlo simulations[3]. We find that the improved spatial resolution allows us to distinguish CL emission peaks from grain interiors and grain boundaries in CdTe lamellae. The CdTe lamellae were prepared from CdTe solar cells using focus-ion-beam milling with a wedge shape to provide a controlled variation in sample thickness. Both a spectral redshift ($\approx 0.01 \pm 0.0015$ eV) in the CL peak location and a reduction in intensity are observed at the grain boundaries with respect to that of the grain interiors (Figs. 1a and b), suggesting an increased concentration of non-radiative recombination centers at the grain boundaries. Energy-dispersive X-ray spectroscopy (EDX) data showed chlorine – a dopant that improves CdTe solar cell performance – segregated at the grain boundaries (Fig. 1c), which may be responsible for the shallow defect states that lead to the spectral redshift. A systematic set of CL measurements with varying lamellar thickness (Fig. 2a) and electron beam current (Fig. 2b) have been performed to build numerical models that can be used to evaluate the contributions from non-radiative surface recombination and radiative bulk recombination. Such information will enable the estimation of the grain boundary recombination velocity based on decreased CL intensities at the grain boundaries.

In summary, TEM-CL enables the room-temperature measurement of CL spectra with high spatial resolution which can be used to compare the radiative recombination rates in grain interiors and at grain boundaries. Combining systematic CL measurements and numerical models, the grain boundary recombination velocity can be estimated. Such information is essential in providing guidelines to improve solar cell efficiency.

^{1.} Center for Nanoscale Science and Technology, National Institute of Standards and Technology, Gaithersburg, Maryland 20899 (USA)

² Maryland NanoCenter, University of Maryland, College Park, MD 20742 (USA)

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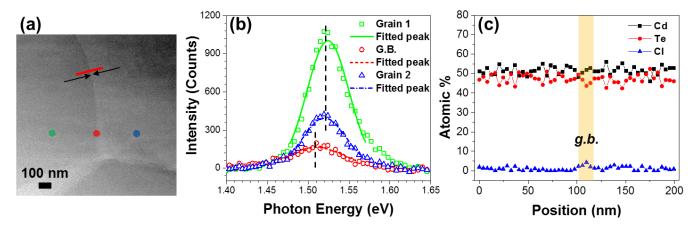


Figure 1. (a) Annular dark-field (ADF) image of a CdTe lamella. (b) CL spectra of grains 1 (green), 2 (blue) and the grain boundary in between (red) as marked by dots in (a). (c) EDX line-scan showing the elemental distributions of Cd, Te and Cl.

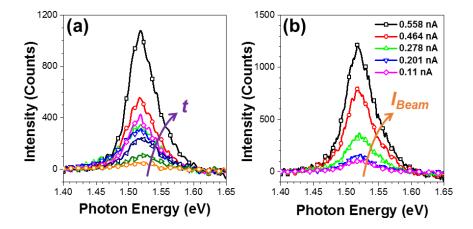


Figure 2. CL peak intensities as functions of (a) sample lamella thickness (t), and (b) electron beam current (I_{Beam}).