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The role of transition radiation in cathodoluminescence imaging and spectroscopy of thin-foils

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ABSTRACT

There is renewed interest in cathodoluminescence (CL) in the transmission electron microscope, since it can be combined with low energy loss spectroscopy measurements and can also be used to probe defects, such as grain boundaries and dislocations, at high spatial resolution. Transition radiation (TR), which is emitted when the incident electron crosses the vacuum-specimen interface, is however an important artefact that has received very little attention. The importance of TR is demonstrated on a wedge shaped CdTe specimen of varying thickness. For small specimen thicknesses (< 250 nm) grain boundaries are not visible in the panchromatic CL image. Grain boundary contrast is produced by electron–hole recombination within the foil, and a large fraction of that light is lost to multiple-beam interference, so that thicker specimens are required before the grain boundary signal is above the TR background. This is undesirable for high spatial resolution. Furthermore, the CL spectrum contains additional features due to TR which are not part of the 'bulk' specimen. Strategies to minimise the effects of TR are also discussed.

1. Introduction

Cathodoluminescence (CL) in the electron microscope is a powerful technique for probing radiative transitions in dielectrics [1], plasmons [2,3], defect properties [4–6], strain [7] and carrier lifetime [8–10] at high spatial resolution. CL is typically implemented in a scanning electron microscope (SEM), but recently there has been renewed interest in its application as a transmission electron microscope (TEM) technique. There are several reasons for this. The first is that CL can be combined with electron energy loss spectroscopy (EELS) in the TEM, which measures both radiative and non-radiative energy loss events and is therefore complementary to CL [2]. Advances in monochromation have also enabled the detection of the EELS signal at deep infrared wavelengths [11,12]. The second reason is that for incoherent luminescence (i.e. light emitted by electron–hole pair recombination) the spatial resolution of TEM-CL is superior to SEM-CL. This is due to reduced elastic scattering of the high energy electron beam within a thin-foil compared to a bulk specimen. The higher spatial resolution is ideal for analysis of nanostructures (e.g. quantum wells) as well as atomic-scale defects (e.g. dislocations, grain boundaries).

Here we have imaged grain boundaries in CdTe using CL in both the SEM and TEM. The initial motivation was to examine the role of surface recombination on CL imaging, especially for TEM-CL, where the effect should be larger due to the thin-foil geometry. It was observed that the grain boundary contrast in TEM-CL was anomalously low, despite Monte Carlo simulations and SEM-CL results predicting otherwise. The discrepancy was shown to be due to transition radiation, i.e. the light emitted when the high energy electron enters and exits the thin-foil [13,14]. The incoherent luminescence, which is the useful signal for grain boundary imaging, is therefore superimposed on a coherent luminescence background due to transition radiation. If the latter is sufficiently large the overall CL image contrast is reduced and the CL spectrum contains additional features. This is similar to the well-known 'Stobbs' factor in TEM phase contrast imaging [15] and 'spurious' Cerenkov losses in EELS spectra [16,17]. Transition radiation from thin foils has been reported previously [18–21], but to our knowledge this is the first demonstration of its importance in high spatial resolution CL imaging and spectroscopy. The results highlight the importance of transition radiation in interpretation of TEM-CL data.

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The paper is organised as follows. In the next section (Section 2) experimental details and computational methods are presented. The latter consist of Monte Carlo methods to simulate the steady state carrier distribution volume for incoherent luminescence as well as calculation of the coherent transition radiation. SEM-CL and TEM-CL experimental data are presented in Section 3 along with simulation results to aid the discussion. SEM-CL is a key part to ruling out surface recombination as the source of the anomalous grain boundary contrast in TEM-CL as well as characterising the emission characteristics of the material. In Section 4 the implications of transition radiation for TEM-CL analysis are discussed along with strategies to minimise its effect. Finally conclusions are presented in Section 5.

2. Experimental and computational details

2.1. Experimental methodology

The sample investigated is a ~2.5 μm thick polycrystalline CdTe layer deposited by close space sublimation [22]. CdTe is an exemplar thin-film photovoltaic material and there is considerable interest in its grain boundary optoelectronic properties. The device consists of the following sequence of layers: glass superstrate, fluorine doped SnO_2, ZnO, CdS and CdTe. There is some inter-diffusion of sulphur from CdS into the CdTe, although the sulphur concentration rapidly decreases to ~5 at% within ~100 nm of the interface [22]. Furthermore, the sample has undergone a $CdCl_2$ activation treatment to improve the device efficiency [22]. The CdTe is effectively chlorine doped during this process. Flat samples for SEM-CL were prepared by Ar broad ion-beam polishing a small section (2 × 1 mm) of the device at 1 keV ion energy and 2° incident angle. The sample was examined at room temperature in plan-view using a Hitachi SU70 FEG SEM equipped with a Gatan MonoCL system. A photomultiplier tube was used for panchromatic CL imaging. The dark signal was determined by blanking the beam and was subtracted from the panchromatic CL images. A Faraday cup was used to measure the beam current.

A TEM wedge shape sample (wedge angle 2.5°) was prepared using an FEI Helios 600 focussed ion-beam (FIB) microscope. A wedge geometry allows examination of a range of thicknesses within the same specimen. Final thinning of the specimen was done at 5 keV ion-beam energy to minimise Ga-beam damage. The thickness along the specimen wedge was measured using EELS in the Durham JEOL 2100 F FEG TEM operating at 200 kV. Convergent beam electron diffraction (CBED) was used to measure the absolute specimen thickness at a reference point [23] and thereby calibrate the EELS inelastic mean free path. The gradient in the EELS thickness profile was consistent with a 2.5° wedge angle.

The specimen was examined at room temperature in the Brunel JEOL 2100F FEG TEM operating at 80 kV. The microscope is equipped with a Gatan Vulcan CL system, which has two ellipsoidal mirrors positioned above and below the specimen, giving 57% collection efficiency (i.e. 7.2 sr solid angle). The high angle annular dark field (HAADF) signal was acquired simultaneously with the CL signal in scanning TEM (STEM) mode. For incoherent imaging conditions (i.e. large detector inner angle) and non-channelling specimen orientations the HAADF signal is proportional to the specimen thickness [24]. In order to approximate these conditions the camera length was adjusted so that the HAADF detector inner angle was nearly six times as large as the STEM probe semi-convergence angle. The CL signal was evaluated as a function of the HAADF intensity, which is an indirect measure of the specimen thickness. A photomultiplier tube, operated in pulse counting mode, and CCD camera was used for acquiring CL panchromatic images and spectrum images respectively.

2.2. Computational details

The Monte Carlo method [25] was used to simulate the excess minority carrier distribution volume due to the electron beam. Radiative recombination of electron–hole pairs within this volume gives rise to the incoherent luminescence that produces grain boundary contrast in CL images. The first stage of the simulation involves calculating the electron–hole pair generation function of the incident electron beam. A screened Rutherford cross-section was used for elastic scattering, while inelastic scattering was modelled using a modified Bethe stopping power that is also valid for small electron energies [25]. The *average* electron–hole pair energy for CdTe was 4.65 eV [26] and carrier generation was assumed to take place uniformly along the trajectory segment of the incident electron. Trajectories for 10^5 electrons incident normal to the specimen surface were simulated in two dimensions for statistically significant results. The convergence angle and diameter of the probe were not taken into account, since these have only a secondary effect on the carrier distribution volume, which is governed primarily by the minority carrier diffusion length in CdTe (see below).

From the simulated electron–hole pair generation function (g) the time evolution of the carriers (n) was modelled via the continuity equation [27]:

$$\frac{\partial n(\mathbf{r}, t)}{\partial t} = D\nabla^2 n(\mathbf{r}, t) + g(\mathbf{r}) - \frac{n(\mathbf{r}, t)}{\tau}. \quad (1)$$

where ∇^2 is the Laplacian. The dependence of the terms on the spatial coordinate vector $\mathbf{r}$ and time t is indicated. The minority (i.e. electron) carrier lifetime (τ) was assumed to be 1 ns [28], while a value of 8.28 cm^2/s was used for D [29]. The latter is based on electrical measurements of CdTe thin-film photovoltaic devices [29]. For these conditions the carrier diffusion length ($L=(D\tau)^{1/2}$) is 910 nm. In our measurements the CL acquisition time per pixel was many micro-seconds, i.e. considerably longer than the lifetime, so that steady-state conditions were established and $\partial n/\partial t=0$. Steady-state was numerically approximated by calculating the time evolution of carriers (via Eq. (1) using finite difference methods) for a total time of three lifetimes.

Free surfaces in the specimen impose boundary conditions which must be satisfied at all times, i.e.:

$$S_{sur} n_{sur} = \left| D \frac{\partial n}{\partial z} \right|_{z=0} \cdots \quad (2)$$

n_{sur} is the carrier concentration at the surface and z is the spatial coordinate along the surface normal (the surface is at $z=0$). S_{sur} is the surface recombination velocity. The left hand side of Eq. (2) is the surface recombination rate [27], while the right hand side is the carrier flux diffusing towards the surface to replenish carriers lost to recombination. For a TEM thin-foil Eq. (2) must be applied to both the beam entrance and exit surfaces. A value of 10^5 cm/s was used for the surface recombination velocity [27]. Some simulations also included a grain boundary; the boundary condition at the grain boundary is physically equivalent to a free surface, apart from the fact that carriers can now diffuse from the two neighbouring grains towards the grain boundary (cf. Eq. (2)). The ratio of grain boundary to surface recombination velocity was assumed to be 0.5, 0.25 or 0.1 (see Section 3.1).