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Defects Density of States Model of Cathodoluminescent Intensity and Spectra of Disordered SiO_2

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Abstract

Electron beam measurements have shown that disordered SiO_2 exhibits luminescent behavior, which varies with incident beam energy and current density, sample temperature (T), exposure time, and wavelength. A simple model based on the electronic band structure and defect density of states—used to explain electron transport in highly disordered insulating materials—has been extended to predict the relative cathodoluminescent intensity and spectral radiance for disordered SiO_2 as a function of these variables.

Insulating SiO_2 has a band gap of ~8.9 eV. Hence thermal excitation from the valence to conduction band is highly improbable; excitation is through collisions of the incident high energy electrons. For visible and near-IR (NIR) light to be emitted, there must be other states within the forbidden band gap for electrons to occupy. These localized defect or "trap" states of disordered SiO_2 are due to structural defects or to substitutional chemical defects. The data were fit with the proposed model using saturation dose rate and mean shallow trap energy as fitting parameters, which can be compared with results from independent experiments.

Tests were conducted on two types of disordered SiO_2 samples: (i) thin (~60-200 nm) coatings on reflective metal substrates and (ii) ~80 μm thick bulk samples, much thicker than the electron penetration depth at the energies studied. Luminescence was measured using a visible range SLR CCD still camera, a VIS/NIR image-intensified video camera, a NIR video camera, and a UV/VIS spectrometer, each with NIST-traceable absolute calibration. Sample temperature was varied from ~295 K to 40 K.

The overall cathodoluminescence intensity depended on the incident electron energy and sample thickness, as predicted by the model. Each incident energy has a corresponding penetration depth, or range, which determines the fraction of energy absorbed in the material. In the thinner samples, the range exceeded the thickness of the sample; therefore, the intensity decreases with increasing energy. However, for the thicker samples, the range is less than the sample thickness and the intensity increased linearly with incident energy.

Cathodoluminescence intensity increased with incident current. At low current densities, luminescent intensity is linearly proportional to incident current density through the dose rate. At very high current densities, saturation can occur when trap states are filled, limiting the number of available states electrons can decay into. Thus, as current increased, the intensity increased until it reached a saturation dose rate on the order of 10^2 Gy/s for disordered SiO_2.

The overall luminescent intensity increased exponentially as T decreased, until it reached a crossover temperature (~45 K), where it began to decrease (as the model predicts). However, the spectral radiance showed there are four distinct bands, corresponding to four distinct energy distributions of defect states (defect bands) within the band gap; each behaved differently with temperature. The UV, blue and green band intensities increases with increasing T, while the red band intensity decreases. The red peak undergoes a red-shift in band peak wavelength as T increases, while the UV, blue and green bands undergo a blue-shift. Each of these relative temperature responses is indicative of the extent to which each band is filled. At low temperature the higher energy defect bands will be largely unoccupied, while those bands below the effective Fermi level will be mostly filled. As T increases, thermal energy excites electrons in lower bands into higher states, thus enhancing the probability for relaxation of conduction band and shallow trapped electrons into the lower bands and the emission of higher energy photons.

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