Multiple exciton generation (MEG) in nano-scale quantum dots (QD’s) represents one of the most important candidate solutions to the fundamental problem of solar energy conversion efficiency in photovoltaic devices. The implication that the efficiency of a single junction cell energy conversion efficiency in photovoltaic devices. The implication that the efficiency of a single junction cell excitation energy are usually correlated with a sudden increase in the QY when hv overlaps the silicon nanocrystal CP, and E2 CP’s (indicated by the dashed vertical lines in the inset) where there is a high joint density of states [9]. We will show that the position of these peaks (and consequently the phonon dispersion around the CP energies) is relatively insensitive to changes in the silicon nanocrystal size, despite evident changes in the principal emission energy (band-gap). We therefore propose an alternative model, to that of MEG here, where enhanced absorption combined with efficient exciton generation occurs when the exciting photon energy, hv matches one of the direct gap transitions in the Si-nanocrystals. We will discuss how defect-like levels, deep within the host SiO$_2$ band-gap, can mediate such excitations and propose simple experiments to elucidate our model. We will also discuss the implications of similar peaks observed in the PLE detected at different emission energies, including for the 1.5µm emission in erbium co-doped films.


Figure 1 Semi-log scale of the Si-nanocrystal PLE spectrum (red circles) detected ~1.4eV with exponential baseline fit (dashed red line). Inset: linear scale of the PLE – baseline difference spectrum. The dashed vertical lines indicate the position of the bulk c-Si $E_1$ and $E_2$ critical points.

Figure 1 shows a typical PLE spectrum from a silicon nanocrystal film (with an emission peak ~885nm). The peaks in the spectrum, which are more easily distinguished after removal of the exponential baseline (inset), are assigned to strong optical absorption and efficient exciton generation when hv overlaps the silicon nanocrystal $E_1$ and $E_2$ CP’s, indicating that these peaks are not due to MEG but due to strong optical absorption and efficient exciton generation when hv overlaps the silicon nanocrystal $E_1$ and $E_2$ CP’s. This effect is observed when the exciting photon energy, hv matches one of the direct gap transitions in the Si-nanocrystals. We will discuss how defect-like levels, deep within the host SiO$_2$ band-gap, can mediate such excitations and propose simple experiments to elucidate our model. We will also discuss the implications of similar peaks observed in the PLE detected at different emission energies, including for the 1.5µm emission in erbium co-doped films.

Evidence for MEG is usually derived from the observation of a fast transient in the absorption, or luminescence, dynamics during high energy (hv $> E_g$) photo-excitation at relatively low intensity, such that the average photo-generated exciton per QD, $<N> <<1$ [3]. This fast transient, on the ps timescale, corresponds to the bi- (or indeed multi-) exciton lifetime, as measured during high intensity excitation ($<N> >>1$). This fast transient is usually correlated with a sudden increase in the QY when hv exceeds the threshold for MEG ($> 2E_g$) with the anticipation of a ‘step-like’ increase in the QY and an increase in the contribution of the fast component to the total absorption (or luminescence decay) transient for hv $= nE_g (n = 2, 3, 4, \ldots)$. However, in spite of the observed ‘step-like’ increase in the QY reported in [1], correlated changes in the fast absorption transient with excitation energy are conspicuous by their absence [1, 5]. Furthermore, the overlapping position of these ‘steps’ in the QY with peaks in the PLE data we have taken, shown in Figure 1.