LOCAL INHOMOGENEITIES IN OPTOELECTRONIC PROPERTIES OF $Cu(In,Ga)(S,Se)_2$ CHALCOPYRIT ABSORBERS BY LATERALLY RESOLVED PHOTOLUMINESCENCE

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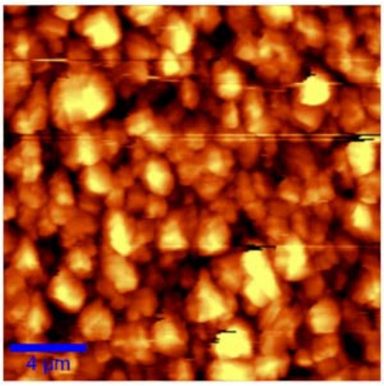
On the basis of light emitted from a volume element with photon flux $\Gamma(\omega)$ being formulated via Planck's generalized law:

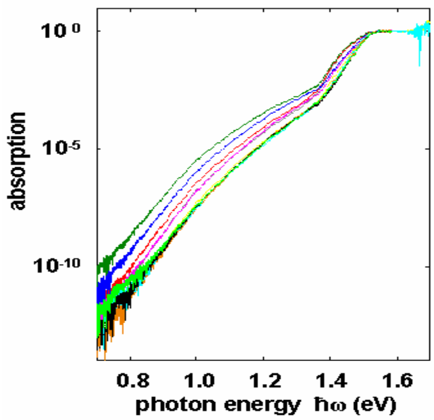
$$\Gamma(\omega) = C\mathrm{E}(\omega)\omega^2 \left[\exp[\frac{\hbar\omega - \mu_{phot}}{kT}] - 1 \right]^{-1}, C = \frac{2\Omega}{h^3 c^2}$$

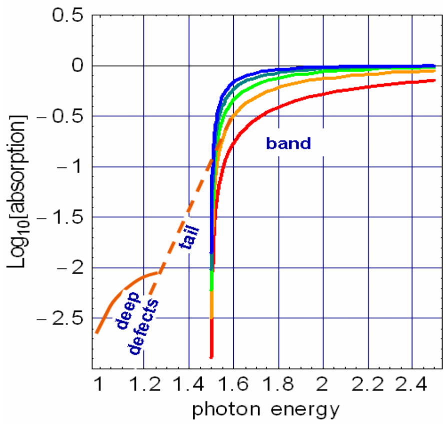
, where Ω, h, c, $E(\omega)$ and μ_{phot} represent solid angle, Planck's constant, speed of light, spectral emissivity and chemical potential of the photon field as well as light attenuated in spectral transmission/reflection/absorption studies we extract the chemical potential of the electron hole system μnp which reads in splitting of quasi-Fermi levels (E_{Fn}-E_{Fp}) and we determine the absorption $A(\hbar\omega) = 1 - \exp[-\alpha(\hbar\omega)\, d]$ of 300K illuminated $Cu(In,Ga)S_2$ absorbers with lateral resolution ($\Delta x \leq 0.9$ μm). Fig. 1 shows an AFM-topology scan of a typical CIGS absorber ($d_{nominal}$= 2 μm) with rms of the height/thickness fluctuations of > 250 nm.



***Figs. 1a**: AFM-scan for demonstrating thickness fluctuations (surface roughness) of a typical polycrystalline $Cu(In,Ga)S_2$ absorber (d_{CIGS2} =2 μm).*

Due to the grainy structure those polycrystalline absorber show lateral variation in optical and optoelectronic properties like absorption and splitting of quasi-Fermi levels. Exemplarily some locally resolved absorptions are shown in fig 2. For the evaluation of the experimental data we artificially compose the absorption by that of an ideal direct semiconductor ($\alpha(\hbar\omega) \sim \{(2m_c)^{3/2}/(2\pi^2\hbar^3)\}\ (\hbar\omega\text{-}E_g)^{1/2}$ degraded by an additional exponentially decaying band tail absorption (stress, strain, limited geometrical volume) and a sub gap absorption induced by defect states somewhat deep in the gap (see fig. 3)

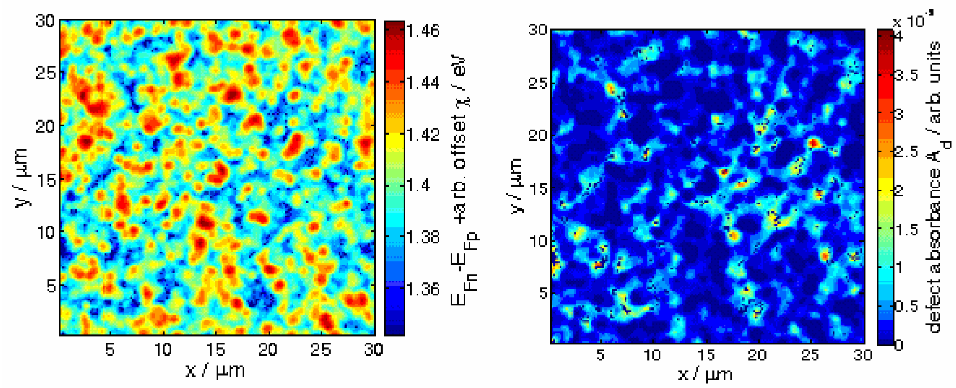




***Figs. 2, 3**: Experimentally detected local absorptions exemplarily plotted for 5 different positions (lateral resolution ≤ 1μm) (left) and artificial assembly of absorption of an ideal direct semiconductor degraded additionally by contributions from tail and deep gap states (right).*

We extract from the lateral scans of the spectral transmission an optical threshold energy, the contribution of the band tails to the absorption $A_{tail}(x,y)$), and that of the deep subgap states $A_{Def}(x,y)$, shown in Fig.4. Furthermore from the high photon energy wing of the spectral luminescence we determine lateral fluctuations of the quasi-Fermi levels (E_{Fn}-E_{Fp}) via Planck's generalized law depicted in Fig. 5.

The correlation of these magnitudes, here exemplarily $A_{Def}(x,y)$ with $\Delta[E_{Fn}-E_{Fp}](x,y)$ shows strong anti-correlation (for the entire scan area $C_{corr} < -0.7$) which convincingly points towards considerable recombination of photogenerated minorities (electrons) via subgap defect states located at about about 300 meV below the conduction band.



Figs. 4,5: *Lateral scans of fluctuation in quasi-Fermi level splitting (left) and of defect absorption A_{Def} resulting from sub gap absorption (right). .*

The analytical formulation of this interlink between $\Delta[E_{Fn}-E_{Fp}]$ and A_{Def} bases upon the relation between high photon energy PL-wing, where the absorption approaches unity A -->1:

$Y_{PL,i}(\omega(A_i->1) \Rightarrow (E_{Fn,i} - E_{Fp,i})$ with the approximation for the position of the minority quasi-Fermi level $E_{Fn,i} = kT\ln[\frac{\Delta n_i + n_0}{n_0}] = kT\ln[\frac{\Delta n_i}{n_0} + 1],$ and with minority behavior $\Delta n_i \gg n_0$ of the thermal equilibrium density $n_{0,i}$ and excess density Δn_i .

We formulate the excess density Δn_i and quasi-Fermi level for minorities under photo generation g, with monomolecular recombination kinetics (SRH) for steady state, wth minority lifetime τ_i, and defect density $N_{D,i}$, each at the individual sites i : $\Delta n_i = g\tau_i = g(\frac{C}{N_{D,i}})$

and write $\left(E_{Fn,i}/kT\right) = \ln[gC] - \ln[n_0] - \ln[N_{D,i}] = C_0 - \ln[N_{D,i}]$ from which we finally see the anticorrelation between local defect density $N_{D,i}$ and local fluctuation of the minority quasi Fermi level $\Delta E_{Fn,i}$ to yield

$$\left(E_{Fn,i}/kT\right) - C_0 = \Delta\left(E_{Fn,i}/kT\right) = -\ln[N_{D,i}]$$