Identifying recombination mechanisms in perovskite solar cells using transient ideality factors

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The ideality factor of a solar cell, derived from the open circuit voltage V_{OC} dependence on light intensity ϕ, has classically been used to identify the dominant recombination mechanism in photovoltaic devices. However, applying this 'Suns-V_{OC}' technique to perovskite cells is problematic since open circuit voltage values change with time owing to the presence of mobile ionic charge in the perovskite layer. This V_{OC} evolution depends on the previously applied bias V_{pre}, the light intensity, and the device architecture. Here we show that the dominant recombination mechanism in different perovskite device architectures (Figs. 1a and 1b) can be identified from the signature of the transient ideality factor $n_{id}(t)$ following application of a forward bias in the dark. $n_{id}(t)$ is measured by monitoring the evolution of the open circuit voltage as a function of time at different light intensities (Fig. 1c). The ideality factor is then extracted using the slope of the V_{OC} versus $\log(\phi)$ plot at each point in time throughout the measurement (see examples in Fig. 1d).

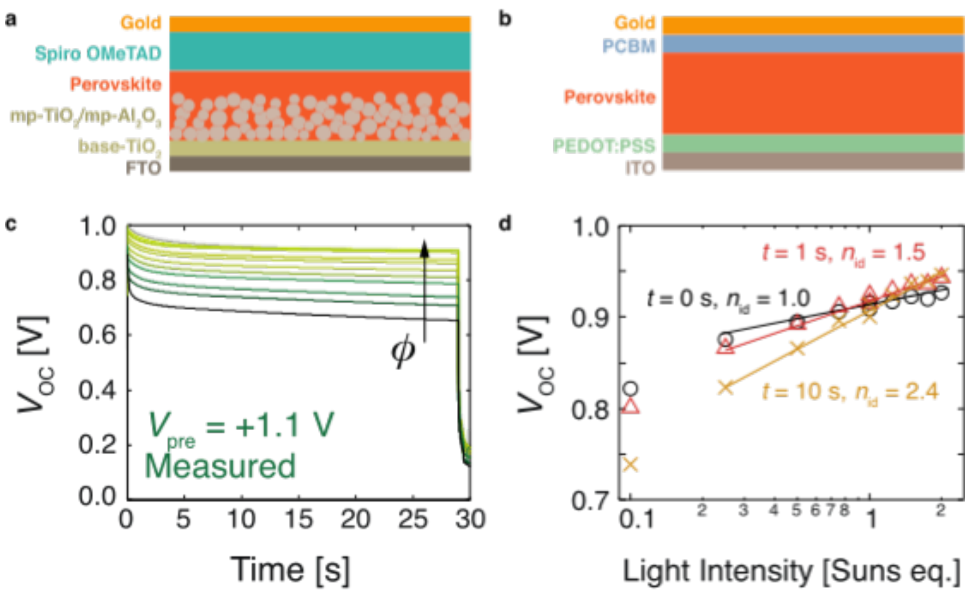

Figure 1. Architectures of perovskite solar cell and example transient Suns-V_{OC} measurement. (**a**) Standard orientation mesoporous (mp) and (**b**) Inverted orientation perovskite device stacks. (**c**) Open circuit voltage evolution for increasing light intensity ϕ for a standard mp-Al_2O_3 perovskite device after preconditioning in the dark for 60 s with a forward bias of $V_{pre} = +1.1$ V. (**d**) Transient ideality factor n_{id}, inferred from Suns-V_{OC} in panel **c** at $t = 0$ s (black circles), $t = 1$ s (red triangles), and $t = 10$ s (gold crosses).

Using a one-dimensional drift-diffusion model we investigate the effect of mobile ionic charge carriers on the transient ideality factor of simulated *p-i-n* devices with a range of recombination schemes. We show that both a change in the overlap of electron and hole populations (Figs. 2a – 2d) and the light intensity-dependence of mobile ion distributions within devices can result in changes in the measured ideality. Consequently, ion migration can account for the transient ideality factors observed in perovskite devices: a change in the dominant recombination process is not required.

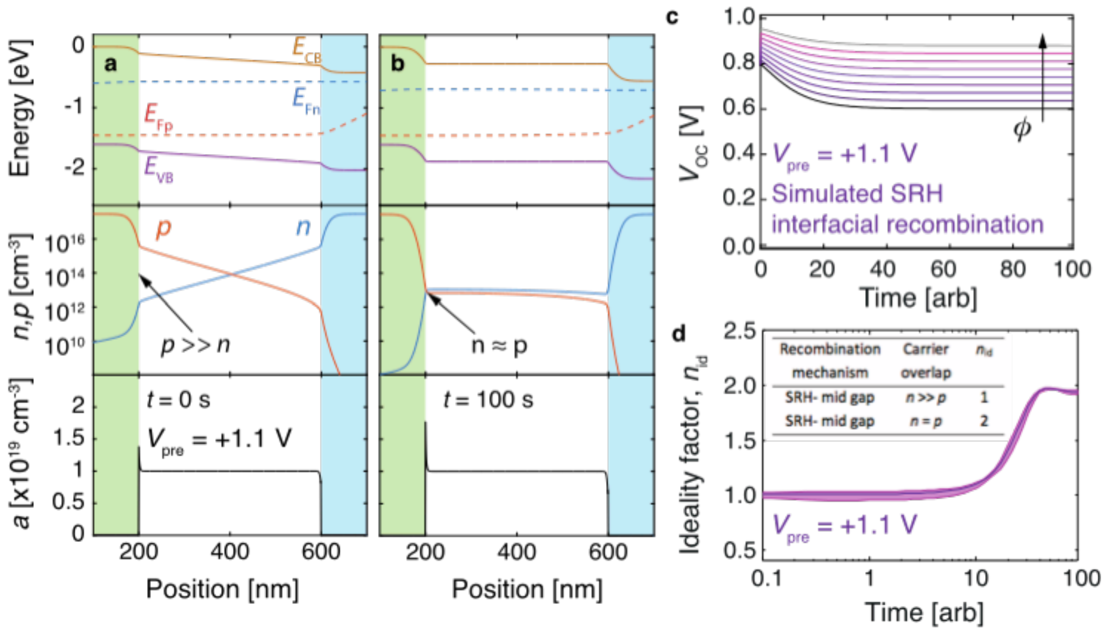

Figure 2. Simulated Energy level diagram, charge densities and transient V_{OC} evolution for a *p-i-n* device at open circuit. (**a**) Energy level diagram, electron *n*, hole *p* and mobile ionic charge *a* densities for a simulated device dominated by SRH recombination at the interfaces after preconditioning at V_{pre} = +1.1 V and being switched to open circuit. (**c**) Simulated open circuit voltage transients following illumination at increasing light intensity for the same device. (**d**) The transient ideality factor changes from 1 to 2 following preconditioning at V_{pre} = +1.1 V due to the change in charge carrier overlap consistent with zero-dimensional theoretical predictions (inset table).

Following this analysis we show that the signature evolution of the ideality after forward biasing can be correlated to theoretical predictions for different recombination types (Fig. 3). The results show that a standard architecture $CH_3NH_3PbI_3$ device with an inert mesoporous (mp) Al_2O_3 scaffold is dominated by interfacial recombination through deep level trap states, whereas a similar device employing a mp-TiO_2 scaffold is dominated by recombination throughout the mesoporous region. An inverted device shows a transient ideality signature consistent with recombination through intermediate trap states. This work provides a powerful new diagnostics tool for identifying the dominant recombination mechanism in perovskite devices.

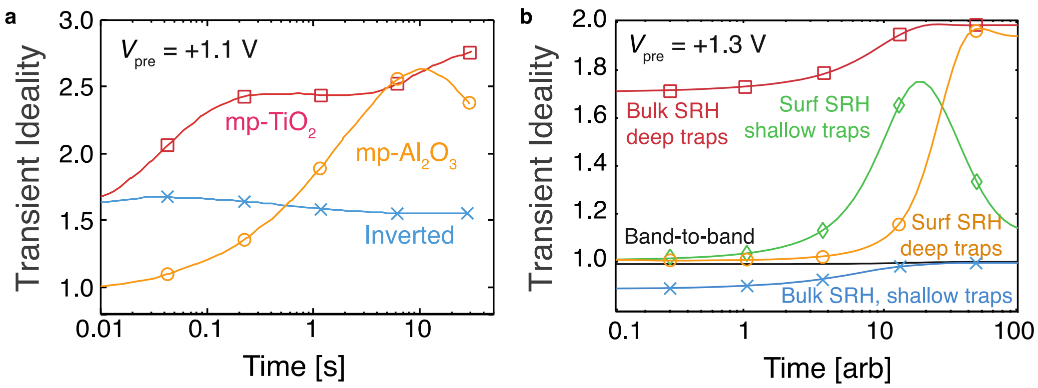

Figure 3. Experimental measurements and simulations of transient ideality factors on perovskite solar cells. (**a**) Experimental transient idealities after prebiasing at V_{pre} = +1.1 V for 60 s for three different architectures of perovskite device. (**b**) Simulated transient ideality factor after preconditioning at V_{pre} = +1.3 V for a *p-i-n* device with mobile ions and different recombination schemes. The evolution of $n_{id}(t)$ provides a signature for different recombination schemes in perovskite devices.