Supporting Information

S1. Magnification of Figure 10 from the Main Article

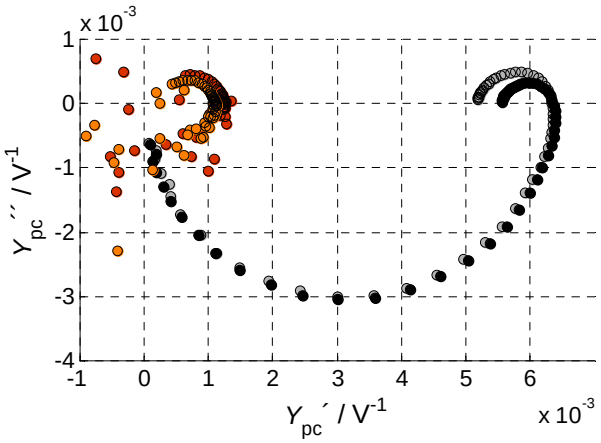

Figure S1: IMPS measurements conducted with the white and orange LEDs (magnification of **Fehler! Verweisquelle konnte nicht gefunden werden.** from the main article).

S2. Fits of IMPS Data (withe LED, 4300 K)

Figure S2 shows the IMPS measurements with the white LED. These were fitted with the equivalent circuit model (ECM) shown in Figure 7 in the main article, which consists of a series resistance and two RQ elements. The fits are also shown in Figure S2.

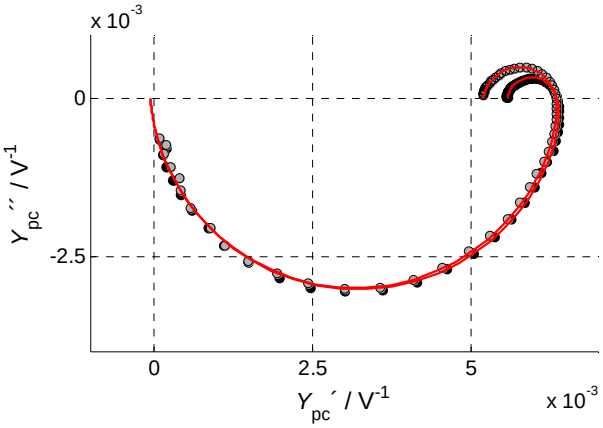

Figure S2: IMPS measurements at 1.35 VRHE with the white LED (4300 K) for 50 mW/cm² (grey symbols) and 100 mW/cm² (black symbols) bias light intensities. Obtained fits are also shown (red lines).

Figure S3 shows the residuals of the fits in Figure S2. Apart from some deviations at high frequencies, which are due to measurement noise, the fits show very small residuals.

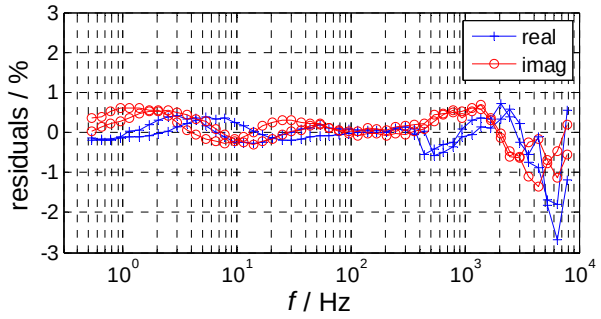

Figure S3: Residuals of both fits shown in Figure S2.

S3. Fits of IMPS Data (orange LED, 590 nm)

Figure S4 shows the IMPS measurements with the orange LED. These were also fitted with the simple equivalent circuit model consisting of a series resistance and two RQ elements. These fits are also shown in Figure S4.

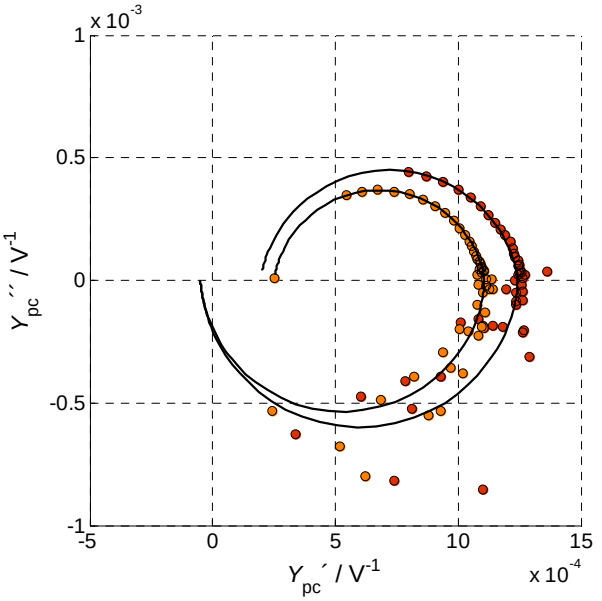

Figure S4: IMPS measurements at 1.35 VRHE with the orange LED (590 nm) for 4.5 mW/cm² (orange symbols) and 9 mW/cm² (red symbols) bias light intensities. Obtained fits are also shown (black lines).

Even though the measurement is very noisy for high frequencies, the relevant information about $Y_{pc}^{+}(0)$ and $Y_{pc}^{-}(0)$ is obtained with good accuracy as can be seen from the residuals of the fits in Figure S5. The deviations from the origin for very high frequencies are very small and of minor relevance for the ratio between charge transfer and recombination. The fact that both fits end up very close rather increases the confidence in these fits.

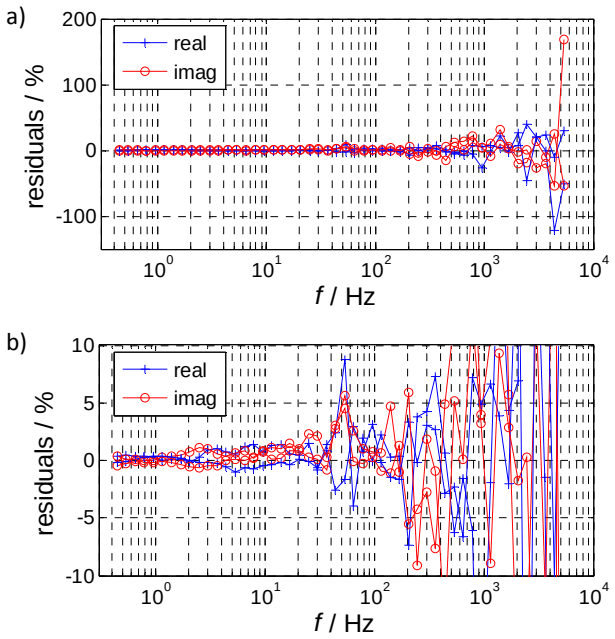

Figure S5: Residuals of both fits shown in Figure S4 in two magnifications.

The residuals in Figure S5 are shown in two magnifications. As expected, the noise in the IMPS measurements at high frequencies causes very large residuals. In fact, these reach values beyond 100% for the highest fitted frequency (5300 Hz). However, starting from the lowest frequencies, the residuals show reasonable values up to 100 Hz, which is already beyond the high frequency intersect (HFI) of the IMPS spectrum on the right end of the spectra.

S3. Correlation Analysis of the Values Obtained for η_t with the Different Methods

A correlation analysis was conducted for the values of η_t obtained by the different methods, as compiled in Table 1 in the main article. The results of this analysis are shown in Table 2 in the main article. The diagrams in Figure S6 show the corresponding fits: chopped light measurements (CLM), the IMPS model approach and hole scavenger measurements (HSM) plotted against the results obtained from the rigorous IMPS analysis. The fits are conducted as a straight line through the origin. The slope of this line represents the expected value for η_t for the respective method with respect to the result from the comprehensive IMPS analysis. Then the standard deviation of these values with respect to the obtained line was calculated. These values are compiled in Table 2 in the main article.

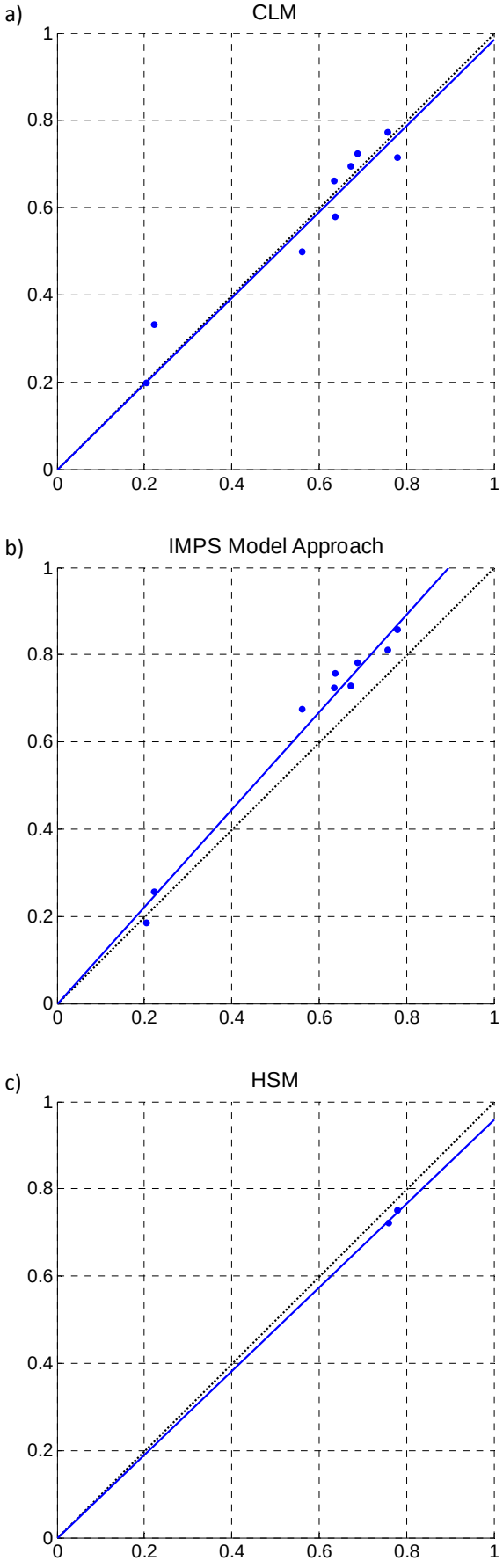

Figure S6 (on the right): Correlation analysis for the results for η_t obtained from the different methods as compiled in Table 1 in the main article. The black dotted line represents the bisection of the first quadrant, the blue dots are the values for η_t as obtained by the respective method plotted against the results from the comprehensive IMPS analysis and the blue line is the fitted line through the origin through the blue dots, respectively.

S4. Rigorous IMPS Analysis for the Coloured LEDs

Figures S7 to S10 show the rigorous IMPS analysis results for the coloured LEDs: green (Figure S7), ultraviolet (Figure S8), blue (Figure S9), and orange (Figure S10).

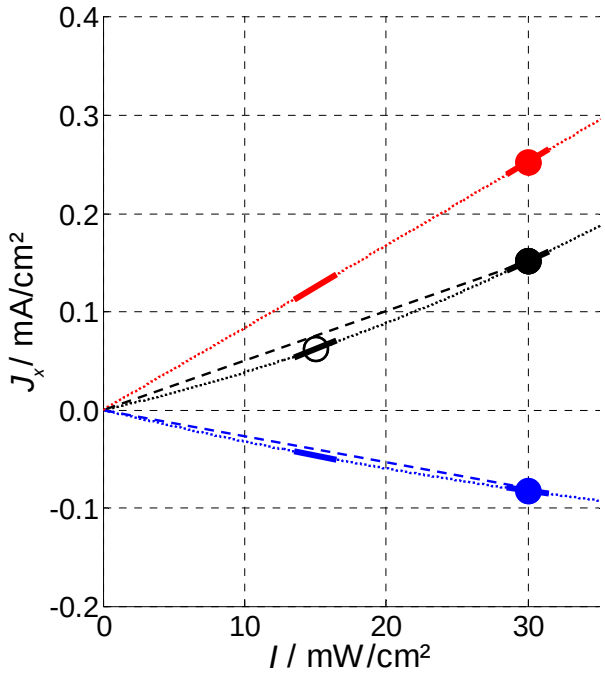

Figure S7: Rigorous IMPS analysis for the green LED at 1.35 V_{RHE}. For a detailed description see Figure 8 in the main article.

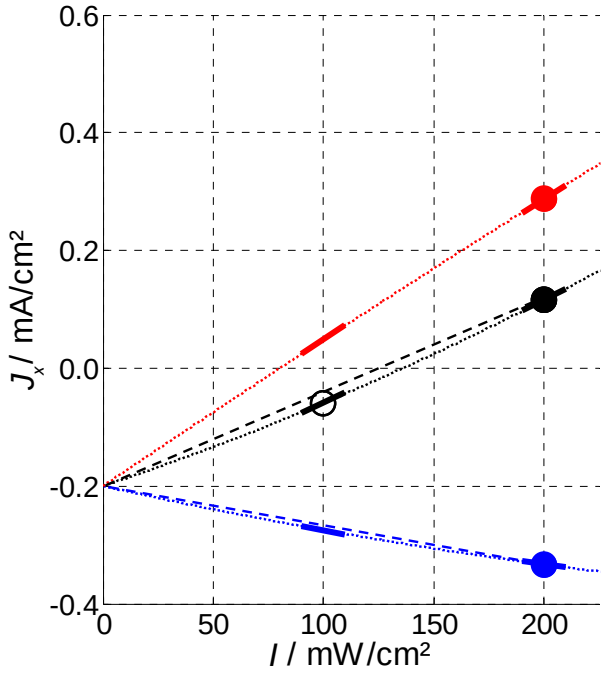

Figure S8: Rigorous IMPS analysis for the ultraviolet LED at 1.35 V_{RHE}. For a detailed description see Figure 8 in the main article.

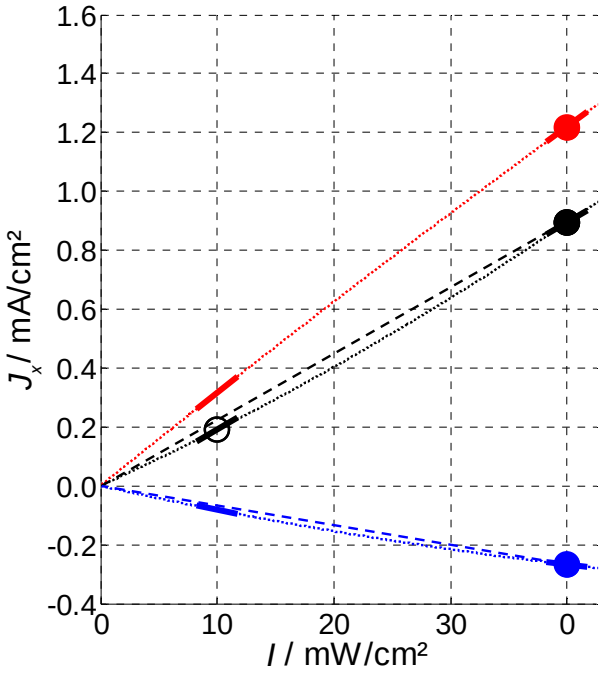

Figure S9: Rigorous IMPS analysis for the blue LED at 1.35 V_{RHE}. For a detailed description see Figure 8 in the main article.

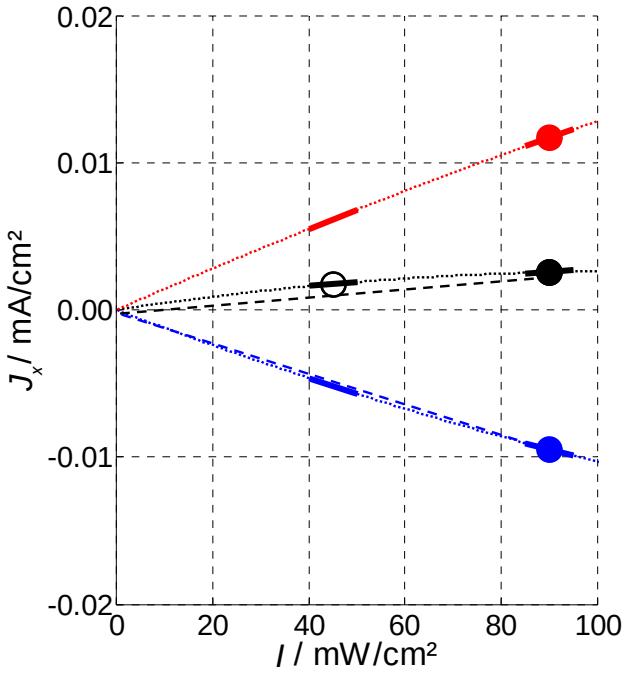

Figure S10: Rigorous IMPS analysis for the orange LED at 1.35 V_{RHE}. For a detailed description see Figure 8 in the main article.