

Correction to “Strong Plasmon Enhancement of the Saturation Photon Count Rate of Single Molecules”

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Recently we published an article in *Journal of Physical Chemistry Letters* titled “Strong Plasmon Enhancement of the Saturation Photon Count Rate of Single Molecules” (DOI: 10.1021/acs.jpcllett.0c00155; publication date: February 19, 2020). Unfortunately, it recently came to our attention that there is a typographical error in the discussion of Figure 6.

Following the comments by one of the referees we modified the manuscript regarding the triplet state modifications, but we mistakenly state that the ratio γ_{isc}/γ_T might be at the origin of the mismatch we find in I_{sat} . Obviously, if PCR_{max} matches the expectation, the ratio γ_{isc}/γ_T also matches the expectation. The text should therefore read that not the ratio γ_{isc}/γ_T but the absolute value of γ_{isc} is at the origin of the mismatch. If possible we would also like broaden the argument and include one sentence that an overestimation of the particle–fluorophore spacing might contribute as well because γ_{nr} scales more strongly with distance than γ_r .

The paragraph containing the above-mentioned additions and corrections should read as follows:

In the simulations we have treated the term $\frac{1 + \gamma_{isc}^0 / \gamma_T^0}{1 + \gamma_{isc} / \gamma_T}$ appearing in both eq 3 and 4 as unity, following Ebbesen et al.¹ Considering the fact that the PCR_{max} enhancement closely follows the prediction this could indicate stronger than expected plasmonic modification of γ_{tot} which contains contributions of γ_{nr} and γ_{isc} . First, a particle–fluorophore spacing smaller than expected by only 0.5 nm causes a 2-fold increase in I_{sat} , whereas PCR_{max} increases by only 10%. This is caused by the fact that γ_{nr} depends more strongly on particle–fluorophore spacing than γ_r . Second, modification of γ_{isc} might also play a role in the higher I_{sat} . Modification of γ_{isc} has also been reported, but experimental studies are limited to a select number of cases^{2–6} that indeed report modest modifications. A quantitative investigation of triplet modifications requires a temporal resolution that is not accessible in our current camera-based setup but could be further investigated using, for example, fluorescence correlation spectroscopy (FCS).^{6–8}

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