# Illuminating the Unexpected: Mechanistic Insights Into Photocatalysis

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Photocatalysis has progressed tremendously over the last decade or so thanks to numerous applications in organic synthesis, where the energy of excited states is used to induce reactions involving high-energy intermediates that would be otherwise inaccessible using conventional non-photoinduced means. Many times, however, the mechanistic details are lacking which can limit the scope and efficiency of the process. Our work centers around investigating the mechanisms of unusual photocatalytic processes such as the upcycling of polymers,[1,2] protecting white wines from the light-struck flavor, or the reduction of atmospheric nitrogen into ammonia.[3] The latter process is particularly important as the anthropogenic production of ammonia from $N_2$ represents a critical necessity to ensure global food security as well as fine and commodity chemical feedstocks.

Coupling nitrogen reduction with water oxidation (Fig. 1) is an attractive approach which may be applied to other photocatalytic processes in water. Our results show that it is possible to harness hydrated electrons released from photo-detachment by using a catalyst to afford ammonia. At room temperature, its efficiency is limited by the solubility of nitrogen in water which can be circumvented by the use of medium to high pressures (100 – 200 bar) to reach quantum yields of 10.8 % (i.e. with an efficiency > 80% when considering that three electrons are needed per ammonia molecule and a further three for water oxidation). To facilitate NH3 quantification in water, we have developed a technique based on solid-phase microextraction combined with GC-MS spectrometry.[4]

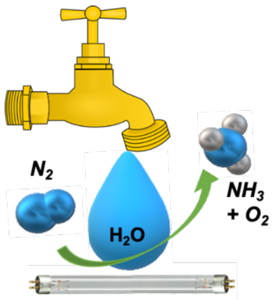


**Fig. 1**. $NH_3$ production from $N_2$.

References

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