

The Dynamics of Organic and Low-Dimensional Inorganic Materials in Photo(Electro) Catalysis

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Abstract

For its potential use in resolving energy and environmental problems, photo(electro)catalytic processes need to be studied at a fundamental level. Less research has been done on the exciton effect, which has an impact on their photo(electro)catalytic performance, particularly in organic and low-dimensional inorganic materials. Investigating the exciton dynamics involved in the photo(electro)catalytic process is therefore crucial, and strategies for regulating exciton dynamics are also required for enhancing photo(electro)catalytic performance. In this study, we outline the principles for controlling exciton dynamics in photo(electro)catalysis and introduce the exciton dynamics and key exciton characterisation tools. Finally, the issues now facing exciton dynamics in the photo(electro)catalytic process are explored, along with potential future advances. We anticipate that this study will shed light on the dynamics of exciton in photo(electro)catalysis, igniting further investigation into materials with potent exciton effects in photo(electro)catalytic applications.

Keywords: *Deterministic behavior; Molecular individuality; Amyloidosis; Prion; Spongiform encephalopathy*

INTRODUCTION

Water splitting and CO₂ reduction are two photo(electro)catalytic applications that have received a lot of interest because they are thought to be viable solutions to the energy crisis and environmental pollution problems. [1,4]. Low-dimensional inorganic materials with an extraordinarily high specific surface area and organic photocatalysts (such as organic conjugated polymers and small molecules) with accessibility, chemical adaptability, and adjustable characteristics have been chosen for diverse photo(electro)catalytic applications.

After light has been absorbed, an exciton is a result of the binding of the electron and the hole. To carry out the redox processes in photo(electro)catalysis, the electron and the hole must be separated [5]. When exposed to light, inorganic bulk photocatalysts produce loosely bound excitons or free charge carriers that have binding energies (0.01 eV) less than the thermal energy at room temperature (kBT, 0.025 eV) and are easily capable of splitting into electrons and holes. Therefore, it is not crucial to consider the exciton dynamics in such inorganic bulk photocatalysts. Contrarily, upon light excitation, stable excitons with relatively high binding energies (typically ranging from 0.25 to 1.2 eV) exist in organic and low-dimensional inorganic materials, owing to the weak intermolecular electronic coupling, low dielectric constant, and reduced dielectric screening in low-dimensional limit of organic materials, as well as their presence in organic materials. Therefore, it is important to carefully evaluate their associated exciton dynamics. The exciton binding energy, or the energy difference between the bound state of an electron and a hole (i.e., exciton), and their fully separated state, can also be used to sort the excitons [6]. This energy difference is described as the strength of the Coulomb interaction between the electron and the hole. The electron is localised in the same or a nearby molecule as the hole in the Frenkel exciton, which has a firmly bound electron-hole pair with a high binding energy and a short Bohr radius (approximately 1 nm). The electron in the Wannier-Mott exciton, in contrast, is not localised close to the hole since it is a weakly coupled electron-hole pair with a large Bohr

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radius (approximately 10 nm) [7].

In general, the photo(electro)catalytic charge transfer process involves three primary physiochemical steps:

- (1) Light absorption under irradiation to produce free charge carriers
- (2) Transport and transfer of free charge carriers to the reaction surface
- (3) Redox reactions at the material/electrolyte interface
- (4) The unwanted free charge recombination may also occur.

While the dynamics of the excitons, rather than free charge carriers, are involved in the photo(electro)catalytic charge transfer process before step (3) for the organic and low-dimensional inorganic materials with strong exciton effect, including exciton generation, diffusion, dissociation, and recombination. A material's ground state (S_0) electrons are stimulated to the singlet excited state upon light excitation, leaving positively charged holes in the S_0 , producing singlet excitons. A triplet exciton is created when a singlet exciton is formed and then through an intersystem crossing (ISC) process to transit to a triplet excited state with an energy lower than that of the singlet excited state. Then, after going through a diffusion process, the singlet and triplet excitons separate into electrons and holes. The exciton dissociation may take place at the interface with an electron or a hole acceptor, in response to interactions with impurities and defect sites, in the presence of an external electric field, or in response to additional thermal energy that is sufficient to outweigh the exciton binding energy. The exciton recombination, which limits the exciton's ability to dissociate and has a negative impact on the efficiency of photo(electro)catalytic reactions, also takes place at the same time. Examples include the radiative decay of the exciton back to its ground state and exciton-exciton annihilation. Additionally, the many-body effect can influence the creation, dissociation, and recombination of the excitons (for example, multiple exciton generation, exciton-exciton annihilation, and exciton-plasmon coupling).

Organic and low-dimensional inorganic materials also contain exciton-based energy transfer in addition to the aforementioned charge transfer. In more detail, dipole-dipole interaction or electron exchange interaction mediates the energy transfer between the excited exciton of catalysts and the ground-state reactant. In polymeric photocatalysts, for instance, the energy transfer between ground-state oxygen molecules and triplet excitons is linked to the production of 1O_2 [8]. The energy transfer and charge transfer typically provide diverse contributions to photo(electro)catalysis because to variances in parameters including energy level, spin configuration, and charge state.

In light of the foregoing descriptions, photo(electro)catalysis is increasingly involving exciton dynamics, which is crucial to organic and low-dimensional inorganic materials with strong exciton effects. But up to now, only a very small number of reviews have compiled research works on it in photo(electro)catalysis that have either focused on one particular material or modulation technique. In this review, we first provide a summary of the methods for characterizing exciton dynamics, then we concentrate on the most recent developments in this field (including exciton generation, diffusion, dissociation, and recombination) and related modulation techniques in photo(electro)catalysis [9]. Finally, we provide a summary and outlook of exciton dynamics in photo(electro)catalysis.

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