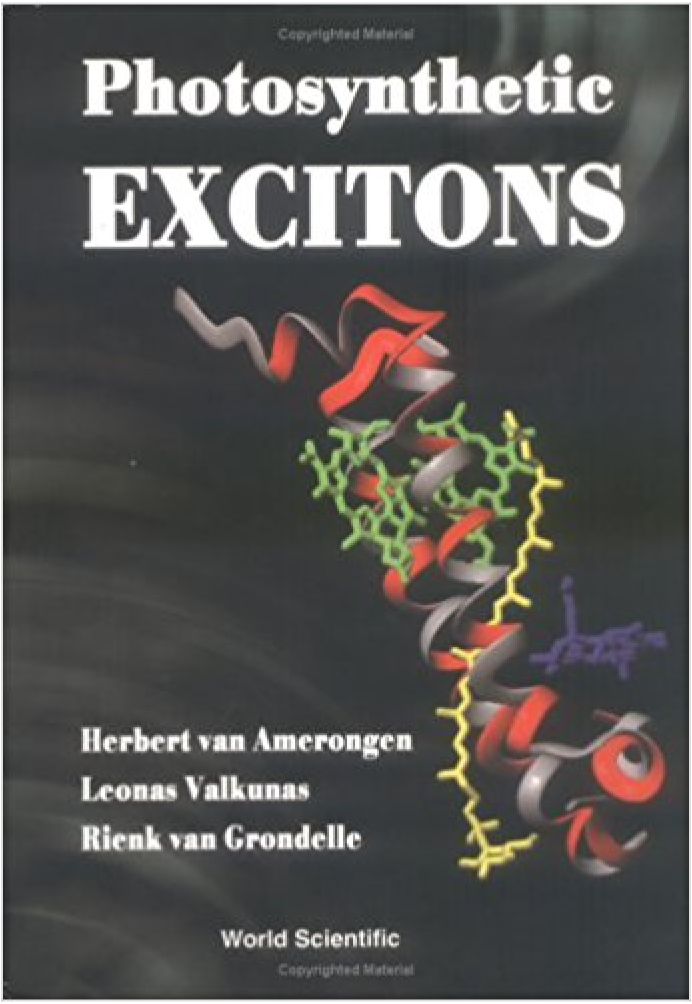
# Photosynthetic Excitons



## Synopsis

Excitons are considered as the basic concept used by describing the spectral properties of photosynthetic pigment-protein complexes and excitation dynamics in photosynthetic light-harvesting antenna and reaction centers. Following the recently obtained structures of a variety of photosynthetic pigment-protein complexes from plants and bacteria our interest in understanding the relation between structure, function and spectroscopy has strongly increased. These data demonstrate a short interpigment distance (of the order of 1 nm or even smaller) and/or a highly symmetric (ring-like) arrangement of pigment molecules in peripheral light-harvesting complexes of photosynthetic bacteria. Books which were devoted to the exciton problem so far mainly considered the spectral properties of molecular crystals. However, the small size of these pigment aggregates in the pigment-protein complexes as well as the role of the protein, which is responsible for the structural arrangement of the complex, clearly will have a dramatic influence on the pigment spectra and exciton dynamics. All these aspects of the problem are considered in this book. Exciton theory is mainly considered for small molecular aggregates (dimers, ring-like structures etc.). Together with the theoretical description of the classical conceptual approach, which mainly deals with polarization properties of the absorption and fluorescence spectra, the nonlinear femtosecond spectroscopy which is widely used for investigations now is also discussed. A large part of the book demonstrates the excitonic effects in a multitude of photosynthetic pigment-protein complexes and how we can understand these properties on the basis of the exciton concept.

## Book Information

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## Customer Reviews

"? this is a unique book ? The price is quite reasonable, and the book should not only be on the shelves of institutional libraries, but also in many private libraries and offices, where it will serve as a standard reference work for many years to come." Photosynthesis Research, 2000

As defined in this book, and in other works on condensed matter physics, an exciton is a superposition of localized molecular excitations. The physics of excitons must of course be described by quantum mechanics (or better yet quantum field theory), and this description is given ample detail in this book for the case of excitons in the photosynthetic system of plants and cyanobacteria. It discusses the physics of photosynthetic excitons by breaking it up into two cases. The first of these is where the broadening the absorption and flourescence bands is larger than the coupling strength between pigments, and hence perturbation theory (the Fermi Golden Rule) can be applied. The second case is where the coupling is the same order of magnitude or greater than the amount of broadening. The first case describes the incoherent energy transfer between the donor and acceptor molecules in the photosynthetic apparatus, and is described by the Forster equation, which was derived early on in the study of the physics of photosynthesis. In the second case one must study `coherent' excitons, which are those where the excitations are not localized on individual molecules.The authors give a comprehensive overview of the dynamics of excitons in photosynthetic complexes and their steady-state spectroscopic properties in this book. Readers of the will probably have different interests when approaching the book, some may want a more theoretical emphasis, while others want to understand the experimental or spectroscopic results in photosynthesis. Both sets of readers will find what they are looking for in the book, and also much more detail if needed in the larger number of references given. All readers are expected to have a solid understanding of the photosynthetic system from a biological or descriptive perspective, and a solid background in quantum physics at the advanced graduate level. The book is fascinating reading, and considering the importance of photosynthesis, is one that certainly become a classic in the field. There is serious discussion at the present time in systems of artificial photosynthesis, and in using the photosynthetic apparatus as a method of computation. Whether this research will get off the ground and produce useful technologies remains to be seen, but certainly an understanding of the physics of photosynthesis as given in this book will play a role in attempting to bring these projects to fruition.Such a long and detailed book cannot be reviewed in the space available, but there are many places in the book that stand out in their clarity and the degree of fascination they instill in the reader and deserve mention. One of these concerns the use of the stochastic Liouville equation in describing the depolarization of a dimer in an ultrafast flourescence or transient absorption experiment. The authors use a `delocalized' representation to calculate the density matrix elements, and consequently calculate the intensity of the flourescence, for orientations of the polarizer both parallel and perpendicular to the excitation and detection branch. Another interesting discussion in the book, because of both its clarity and its conciseness, is one that concerns a kinetic model of a light-harvesting antenna (LHA) complex consisting of several pigments with localized excitons. The LHA is modeled by a homogeneous rectangular or hexagonal lattice. The kinetics within the LHA is then determined by the hopping rate, and the charge separation in the reaction center (RC) is included as a quenching rate on one of the lattice sites. The kinetic model illustrates the role of the LHA, via the dense packing of pigments in it, in enhancing the number of photons that reach the RC. The lifetime of an exciton increases because of this, but only at the expense of the slower decay of excited states. The authors discuss how to determine the relation between the LHA and the RC experimentally by using time-resolved methods to compare the kinetics of the excited states obtained after excitation of the LHA with those obtained by direct excitation of the RC. Particularly interesting in these experiments is the use of genetic mutants lacking LH2 to study the kinetics within the RC. The authors also spend two entire chapters on the nonlinear annihilation of excitons, both from a theoretical and an experimental point of view. The nonlinear processes which they study come primarily from molecules that are in excited singlet (S) or triplet (T) states, and are referred to as singlet-singlet (S-S) and singlet-triplet (S-T) annihilation. They arise primarily at high excitation densities, are diffusion-limited, and result in excess energy being redistributed among the vibrational modes. The authors derive the kinetic equations for a collection of pigment molecules located on the sites of a regular lattice, where each molecule is characterized by a set of singlet states. They then discuss the case where singlet excited molecules can be converted into triplet states via intersystem crossing. The kinetic equations for the density of the excited states are derived, and the authors show how `anti-Smoluchowski triplet-triplet (T-T) annihilation can arise because of the inhomogeneous distribution of triplets resulting from S-T annihilation. They also discuss in some detail the local heating that results from the excitation annihilation. This local heating has the effect of modifying the coupling of the molecule to its surroundings, resulting sometimes in the `polaron effect'. The authors use collective coordinates to study the (linear) coupling between an electron and the collective nuclear motion. The dynamics of the collective coordinates are described by a stochastic damped oscillator equation. For the case where the kinetics of the collective coordinates is fast relative to the changes in temperature and occupation number of the molecule. Thermal fluctuations are thus neglected and the equation can be readily solved. For the case where the collective coordinates change slow with respect to changes in temperature, one again gets a solution, and also a notion of `difference absorption spectrum', which the authors discuss in the context of pump-probe spectroscopy.



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