

University of Groningen

The ultrafast dynamics of aggregate excitons in water

Burgel, Mirjam van; Wiersma, D. A.

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version

Publisher's PDF, also known as Version of record

Publication date:

1999

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Burgel, M. V., & Wiersma, D. A. (1999). *The ultrafast dynamics of aggregate excitons in water*. s.n.

Copyright

Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: <https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment>.

Take-down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): <http://www.rug.nl/research/portal>. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.

**THE ULTRAFAST DYNAMICS OF AGGREGATE
EXCITONS IN WATER**

Beoordelingscommissie:

Prof. dr. R. van Grondelle

Prof. dr. J. Kommandeur

Prof. dr. J. G. Snijders

Ontwerp omslag: Sylvia van den Bosch

This investigation was supported by the Netherlands Foundation for Physical Research (FOM) with financial aid from the Netherlands Organization for the Advancement of Science (NWO).



RIJSUNIVERSITEIT GRONINGEN

**THE ULTRAFAST DYNAMICS OF AGGREGATE
EXCITONS IN WATER**

PROEFSCHRIFT

ter verkrijging van het doctoraat in de
Wiskunde en Natuurwetenschappen
aan de Rijksuniversiteit Groningen
op gezag van de
Rector Magnificus, dr. D.F.J. Bosscher,
in het openbaar te verdedigen op
vrijdag 29 januari 1999
om 16.00 uur

door

Mirjam van Burgel

geboren op 30 januari 1968
te Purmerend

Promotor: Prof. dr. D.A. Wiersma
Co-Promotor: Dr. K. Duppen

Contents

Chapter 1

Introduction	1
1.1 Molecular aggregates	2
1.2 Outline of this thesis	4
References	7

Chapter 2

Experimental techniques	9
2.1 Introduction	10
2.2 The generation of femtosecond pulses	11
2.3 Set-up and detection in different experiments	15
2.4 Excitation characteristics	19
2.5 Sample preparation and handling	22
References	24

Chapter 3

The electronic band structure of one-dimensional aggregates	25
3.1 Introduction	26
3.2 One-dimensional molecular excitons	27
3.3 The exciton delocalization length	33
3.4 Vibronic structure and the Stokes shift	38
References	41

Chapter 4

Exciton dephasing dynamics	43
4.1 Introduction	44
4.2 The stochastic model	45
4.3 Two-pulse photon echo experiments	50
4.4 Chirped four-wave mixing	65
4.5 Exchange narrowing	69
4.6 Summary and conclusions	72
Appendix: Evaluation of the stochastic relaxation functions eqs. (4.20)	74
References	77

Chapter 5

The three-pulse photon echo	79
5.1 Introduction	80
5.2 Three-pulse stimulated photon echo experiments	82
5.3 Theory of the three-pulse stimulated photon echo experiment	86
5.4 Model calculations	95
5.4.1 The stimulated photon echo as a function of delay τ	95
5.4.2 The stimulated photon echo as a function of delay T	99
5.5 Summary and conclusions	102
References	104

Chapter 6

Exciton annihilation

105	
6.1 Introduction	106
6.2 Experiments	107
6.3 Exciton population decay	108
6.4 Diffusion-limited population decay	115
6.5 Monte Carlo simulations	121
6.6 Discussion	122
6.6.1 Annihilation of localized excitations	122
6.6.2 Annihilation of delocalized excitations	126
6.7 Summary and conclusions	128
References	130

Samenvatting	131
--------------	-----

Dankwoord	137
-----------	-----