



Universiteit  
Leiden  
The Netherlands

## Probing spatial heterogeneity in supercooled glycerol and temporal heterogeneity with single-molecule FRET in polyprolines

Xia, T.

### Citation

Xia, T. (2010, March 25). *Probing spatial heterogeneity in supercooled glycerol and temporal heterogeneity with single-molecule FRET in polyprolines*. Casimir PhD Series. Retrieved from <https://hdl.handle.net/1887/15122>

Version: Corrected Publisher's Version

License: [Licence agreement concerning inclusion of doctoral thesis in the Institutional Repository of the University of Leiden](#)

Downloaded from: <https://hdl.handle.net/1887/15122>

**Note:** To cite this publication please use the final published version (if applicable).

Probing spatial heterogeneity in supercooled  
glycerol and temporal heterogeneity with  
single-molecule FRET in polyprolines

PROEFSCHRIFT

ter verkrijging van  
de graad van Doctor aan de Universiteit Leiden,  
op gezag van de Rector Magnificus prof. mr. P. F. van der Heijden,  
volgens besluit van het College voor Promoties  
te verdedigen op donderdag 25 maart 2010  
klokke 11:15 uur

door

**Ted (Tie) Xia**  
geboren te Dalian, China  
in 1977

**Promotiecommissie:**

Promotor: Prof. Dr. M. A. G. J. Orrit  
Overige Leden: Prof. Dr. B. Schuler (Universität Zürich)  
Prof. Dr. J. M. van Ruitenbeek  
Prof. Dr. E. J. J. Groenen  
Prof. Dr. M. van Hecke  
Prof. Dr. T. Schmidt

The presented work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).

Casimir PhD Series, Delft-Leiden, 2010-03  
ISBN: 978-90-8593-068-6

# Contents

<b>1</b>	<b>Introduction</b>	<b>1</b>
1.1	Glass transition and heterogeneity . . . . .	1
1.2	Single-molecule fluorescence and temperature-cycle microscopy	2
1.3	Outline of the thesis . . . . .	5
<b>2</b>	<b>Soft glassy rheology of supercooled liquids</b>	<b>9</b>
2.1	Introduction . . . . .	10
2.2	Experimental methods . . . . .	11
2.2.1	Sample preparation . . . . .	11
2.2.2	Design of the Couette cell . . . . .	12
2.3	Results . . . . .	12
2.3.1	Viscoelastic behavior and yield stress . . . . .	12
2.3.2	Phenomenological model . . . . .	14
2.3.3	Shear thinning and rejuvenation . . . . .	17
2.4	Discussion . . . . .	20
2.5	Appendix . . . . .	24
2.5.1	Viscosities of glycerol and <i>o</i> -terphenyl . . . . .	24
2.5.2	Onset of solid-like behavior in glycerol . . . . .	24
2.5.3	Experimental details . . . . .	26
<b>3</b>	<b>Aging and solidification of supercooled glycerol</b>	<b>29</b>
3.1	Introduction . . . . .	30
3.2	Experimental methods . . . . .	31
3.3	Results . . . . .	32
3.3.1	Viscosity measurements . . . . .	32
3.3.2	Solidification and importance of cooling rate . . . . .	32
3.3.3	Measurements in the plate-plate geometry . . . . .	35
3.4	Discussion . . . . .	39
3.5	Conclusion . . . . .	42
<b>4</b>	<b>Micron-sized structure in a thin glycerol film revealed by fluorescent probes</b>	<b>43</b>
4.1	Introduction . . . . .	44

4.2	Experimental methods . . . . .	45
4.3	Results . . . . .	46
4.4	Discussion . . . . .	52
4.5	Conclusion . . . . .	55
<b>5</b>	<b>Small-angle neutron scattering on supercooled glycerol</b>	<b>57</b>
5.1	Introduction . . . . .	58
5.2	Experimental methods . . . . .	60
5.3	Results and discussion . . . . .	61
5.4	Conclusion . . . . .	66
<b>6</b>	<b>Temperature-cycle microscopy of single-molecule FRET in polyprolines</b>	<b>67</b>
6.1	Introduction . . . . .	68
6.2	Experimental methods . . . . .	72
6.2.1	Sample preparation . . . . .	72
6.2.2	Optical setup . . . . .	73
6.2.3	Heating calibration . . . . .	75
6.2.4	Data analysis . . . . .	76
6.3	Results . . . . .	77
6.4	Discussion . . . . .	85
6.5	Conclusion . . . . .	87
<b>7</b>	<b>Summary</b>	<b>89</b>
7.1	Heterogeneity in supercooled liquids . . . . .	89
7.2	Temperature-cycle microscopy of single-molecule FRET in poly- prolines . . . . .	91
	<b>References</b>	<b>93</b>
	<b>Samenvatting</b>	<b>107</b>
	<b>List of Publications</b>	<b>113</b>
	<b>Curriculum Vitae</b>	<b>115</b>
	<b>Nawoord</b>	<b>117</b>