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Single molecules in soft matter : a study of biomolecular conformation, heterogeneity and plasmon enhanced fluorescence

Yuan, H.

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Author: Yuan, Haifeng

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References

- [1] E. Rhoades, E. Gussakovsky and G. Haran, “Watching proteins fold one molecule at a time”, *Proc. Natl. Acad. Sci. U. S. A.* **100** (2003) 3197–3202.
- [2] X. Zhuang and M. Rief, “Single-molecule folding”, *Curr. Opin. Struc. Biol.* **13** (2003) 88–97.
- [3] S. McKinney, A. Declais, D. Lilley and T. Ha, “Structural dynamics of individual Holliday junctions”, *Nat. Struct. Mol. Biol.* **10** (2003) 93–97.
- [4] J. N. Onuchic, Z. Luthey-Schulten and P. G. Wolynes, “Theory of protein folding: The energy landscape perspective”, *Annu. Rev. Phys. Chem.* **48** (1997) 545–600.
- [5] J. Sabelko, J. Ervin and M. Gruebele, “Observation of strange kinetics in protein folding”, *Proc. Natl. Acad. Sci. U. S. A.* **96** (1999) 6031–6036.
- [6] R. Rudolph, R. Siebendritt, G. Nessler, A. K. Sharma and R. Jaenicke, “Folding of an all-beta protein: independent domain folding in gamma II-crystallin from calf eye lens.”, *Proc. Natl. Acad. Sci. U. S. A.* **87** (1990) 4625–4629.
- [7] L. M. Gloss, B. R. Simler and C. R. Matthews, “Rough energy landscapes in protein folding: dimeric E. coli Trp repressor folds through three parallel channels”, *J. Mol. Biol.* **312** (2001) 1121–1134.
- [8] C. Wright, K. Lindorff-Larsen, L. Randles and J. Clarke, “Parallel protein-unfolding pathways revealed and mapped”, *Nat. Struct. Mol. Biol.* **10** (2003) 658–662.
- [9] S. Ebbinghaus, A. Dhar, D. McDonald and M. Gruebele, “Protein folding stability and dynamics imaged in a living cell”, *Nat. Methods* **7** (2010) 319–323.
- [10] T. Basché, W. E. Moerner, M. Orrit and U. P. Wild, “Single-molecule optical detection, imaging and spectroscopy”, WILEY-VCH Verlag.
- [11] M. Orrit and J. Bernard, “Single pentacene molecules detected by fluorescence excitation in a p-terphenyl crystal.”, *Phys. Rev. Lett.* **65** (1990) 2716–2719.
- [12] T. Ha, T. Enderle, D. F. Ogletree, D. S. Chemla, P. R. Selvin and S. Weiss, “Probing the interaction between two single molecules: fluorescence resonance energy transfer between a single donor and a single acceptor.”, *Proc. Natl. Acad. Sci. U. S. A.* **93** (1996) 6264–8.
- [13] W. E. Moerner and M. Orrit, “Illuminating single molecules in condensed matter”, *Science* **283** (1999) 1670–1676.

-
- [14] A. Gaiduk, P. V. Ruijgrok, M. Yorulmaz and M. Orrit, "Detection limits in photothermal microscopy", *Chem. Sci.* **1** (2010) 343–350.
- [15] P. Kukura, M. Celebrano, A. Renn and V. Sandoghdar, "Single-molecule sensitivity in optical absorption at room temperature", *J. Phys. Chem. Lett.* **1** (2010) 3323–3327.
- [16] S. Chong, W. Min and X. S. Xie, "Ground-state depletion microscopy: Detection sensitivity of single-molecule optical absorption at room temperature", *J. Phys. Chem. Lett.* **1** (2010) 3316–3322.
- [17] M. Celebrano, P. Kukura, A. Renn and V. Sandoghdar, "Single-molecule imaging by optical absorption", *Nature Photon.* **5** (2011) 95–98.
- [18] J. Hofkens and M. B. J. Roeffaers, "MICROSCOPY Single-molecule light absorption", *Nature Photon.* **5** (2011) 80–81.
- [19] K. Kneipp, Y. Wang, H. Kneipp, L. T. Perelman, I. Itzkan, R. R. Dasari and M. S. Feld, "Single molecule detection using surface-enhanced raman scattering (SERS)", *Phys. Rev. Lett.* **78** (1997) 1667–1670.
- [20] S. Nie and S. R. Emory, "Probing single molecules and single nanoparticles by surface-enhanced Raman scattering", *Science* **275** (1997) 1102–1106.
- [21] K. L. Wustholz, C. L. Brosseau, F. Casadio and R. P. Van Duyne, "Surface-enhanced Raman spectroscopy of dyes: from single molecules to the artists' canvas", *Phys. Chem. Chem. Phys.* **11** (2009) 7350–7359.
- [22] A. Ahmed and R. Gordon, "Single molecule directivity enhanced Raman scattering using nanoantennas", *Nano Lett.* **12** (2012) 2625–2630.
- [23] R. Zhang, Y. Zhang, Z. C. Dong, S. Jiang, C. Zhang, L. G. Chen, L. Zhang, Y. Liao, J. Aizpurua, Y. Luo, J. L. Yang and J. G. Hou, "Chemical mapping of a single molecule by plasmon-enhanced Raman scattering", *Nature* **498** (2013) 82–86.
- [24] T. Ha, A. Y. Ting, J. Liang, W. B. Caldwell, A. A. Deniz, D. S. Chemla, P. G. Schultz and S. Weiss, "Single-molecule fluorescence spectroscopy of enzyme conformational dynamics and cleavage mechanism.", *Proc. Natl. Acad. Sci. U. S. A.* **96** (1999) 893–8.
- [25] X. S. Xie and H. P. Lu, "Single-molecule enzymology", *J. Biol. Chem.* **274** (1999) 15 967–15 970.
- [26] J. Müller, J. M. Lupton, A. L. Rogach, J. Feldmann, D. V. Talapin and H. Weller, "Monitoring surface charge migration in the spectral dynamics of single CdSe/CdS nanodot/nanorod heterostructures", *Phys. Rev. B* **72** (2005) 205 339.
- [27] H. Yang, "Detection and characterization of dynamical heterogeneity in an event series using wavelet correlation", *J. Chem. Phys.* **129** (2008) 074 701.

- [28] D. Woll, E. Braeken, A. Deres, F. C. De Schryver, H. Uji-i and J. Hofkens, “Polymers and single molecule fluorescence spectroscopy, what can we learn?”, *Chem. Soc. Rev.* **38** (2009) 313–328.
- [29] Y. Wang and H. P. Lu, “Bunching effect in single-molecule T4 Lysozyme nonequilibrium conformational dynamics under enzymatic reactions”, *J. Phys. Chem. B* **114** (2010) 6669–6674.
- [30] A. Hoffmann and M. T. Woodside, “Signal-pair correlation analysis of single-molecule trajectories”, *Angew. Chem. Int. Ed.* **50** (2011) 12 643–12 646.
- [31] Y. Tian, P. Navarro, B. Kozankiewicz and M. Orrit, “Spectral diffusion of single dibenzoterrylene molecules in 2,3-dimethylantracene”, *ChemPhysChem* **13** (2012) 3510–3515.
- [32] F. Kulzer, T. Xia and M. Orrit, “Single molecules as optical nanoprobe for soft and complex matter”, *Angew. Chem. Int. Ed.* **49** (2010) 854–866.
- [33] L. J. Kaufman, “Heterogeneity in single-molecule observables in the study of supercooled liquids”, *Annu. Rev. Phys. Chem.* **64** (2013) 177–200.
- [34] E. J. Peterman, H. Sosa and W. Moerner, “Single-molecule fluorescence spectroscopy and microscopy of biomolecular motors”, *Annu. Rev. Phys. Chem.* **55** (2004) 79–96.
- [35] H. Hofmann, F. Hillger, S. H. Pfeil, A. Hoffmann, D. Streich, D. Haenni, D. Nettels, E. A. Lipman and B. Schuler, “Single-molecule spectroscopy of protein folding in a chaperonin cage”, *Proc. Natl. Acad. Sci. U. S. A.* (2010).
- [36] W. E. Moerner, “New directions in single-molecule imaging and analysis”, *Proc. Natl. Acad. Sci. U. S. A.* **104** (2007) 12 596–12 602.
- [37] J. R. Lakowicz, “Principles of fluorescence Spectroscopy, Third Edition”, Springer (2006).
- [38] B. Schuler and H. Hofmann, “Single-molecule spectroscopy of protein folding dynamics expanding scope and timescales”, *Curr. Opin. Struc. Biol.* **23** (2013) 36 – 47.
- [39] D. Talaga, W. Lau, H. Roder, J. Tang, Y. Jia, W. DeGrado and R. Hochstrasser, “Dynamics and folding of single two-stranded coiled-coil peptides studied by fluorescent energy transfer confocal microscopy”, *Proc. Natl. Acad. Sci. U. S. A.* **97** (2000) 13 021–13 026.
- [40] D. Nettels, I. V. Gopich, A. Hoffmann and B. Schuler, “Ultrafast dynamics of protein collapse from single-molecule photon statistics”, *Proc. Natl. Acad. Sci. U. S. A.* **104** (2007) 2655–2660.
- [41] H. S. Chung, K. McHale, J. M. Louis and W. A. Eaton, “Single-molecule fluorescence experiments determine protein folding transition path times”, *Science* **335** (2012) 981–984.

- [42] S. Myong, S. Cui, P. V. Cornish, A. Kirchhofer, M. U. Gack, J. U. Jung, K.-P. Hopfner and T. Ha, “Cytosolic viral sensor rig-i is a 5'-triphosphate-dependent translocase on double-stranded rna”, *Science* **323** (2009) 1070–1074.
- [43] H. S. Chung, J. M. Louis and W. A. Eaton, “Experimental determination of upper bound for transition path times in protein folding from single-molecule photon-by-photon trajectories”, *Proc. Natl. Acad. Sci. U. S. A.* **106** (2009) 11 837–11 844.
- [44] S. Hohng, R. Zhou, M. K. Nahas, J. Yu, K. Schulten, D. M. J. Lilley and T. Ha, “Fluorescence-force spectroscopy maps two-dimensional reaction landscape of the Holliday junction”, *Science* **318** (2007) 279–283.
- [45] M. Pirchi, G. Ziv, I. Riven, S. S. Cohen, N. Zohar, Y. Barak and G. Haran, “Single-molecule fluorescence spectroscopy maps the folding landscape of a large protein”, *Nat. Commun.* **2** (2011).
- [46] M. Margittai, J. Widengren, E. Schweinberger, G. F. Schröder, S. Felekyan, E. Haustein, M. König, D. Fasshauer, H. Grubmüller, R. Jahn and C. A. M. Seidel, “Single-molecule fluorescence resonance energy transfer reveals a dynamic equilibrium between closed and open conformations of syntaxin 1”, *Proc. Natl. Acad. Sci. U. S. A.* **100** (2003) 15 516–15 521.
- [47] Y. Gambin, V. VanDelinder, A. C. M. Ferreon, E. A. Lemke, A. Groisman and A. A. Deniz, “Visualizing a one-way protein encounter complex by ultrafast single-molecule mixing”, *Nat. Methods* **8** (2011) 239–U77.
- [48] H. Kim and T. Ha, “Single-molecule nanometry for biological physics”, *Rep. Prog. Phys.* **76** (2013) 016 601.
- [49] Y. Xu, P. Purkayastha and F. Gai, “Nanosecond folding dynamics of a three-stranded beta-sheet”, *J. Am. Chem. Soc.* **128** (2006) 15 836–15 842.
- [50] J. Kubelka, J. Hofrichter and W. A. Eaton, “The protein folding “speed limit”?”, *Curr. Opin. Struc. Biol.* **14** (2004) 76–88.
- [51] R. Zondervan, F. Kulzer, H. van der Meer, J. A. J. M. Disselhorst and M. Orrit, “Laser-driven microsecond temperature cycles analyzed by fluorescence polarization microscopy.”, *Biophys. J.* **90** (2006) 2958–69.
- [52] J. Kubelka, “Time-resolved methods in biophysics. 9. Laser temperature-jump methods for investigating biomolecular dynamics”, *Photochem. Photobiol. Sci.* **8** (2009) 499–512.
- [53] H. Sillescu, “Heterogeneity at the glass transition: a review”, *J. Non-Cryst. Solids* **243** (1999) 81–108.
- [54] M. D. Ediger, “Spatially heterogeneous dynamics in supercooled liquids”, *Annu. Rev. Phys. Chem.* **51** (2000) 99–128.
- [55] P. G. Debenedetti and F. H. Stillinger, “Supercooled liquids and the glass transition”, *Nature* **410** (2001) 259–267.

-
- [56] M. Orrit, “Towards a molecular view of glass heterogeneity”, *Angew. Chem. Int. Ed.* **52** (2013) 163–166.
- [57] L. Deschenes and D. Bout, “Heterogeneous dynamics and domains in supercooled o-terphenyl: A single molecule study”, *J. Phys. Chem. B* **106** (2002) 11 438–11 445.
- [58] R. Zondervan, F. Kulzer, G. C. G. Berkhout and M. Orrit, “Local viscosity of supercooled glycerol near T-g probed by rotational diffusion of ensembles and single dye molecules”, *Proc. Natl. Acad. Sci. USA* **104** (2007) 12 628–12 633.
- [59] S. A. Mackowiak, T. K. Herman and L. J. Kaufman, “Spatial and temporal heterogeneity in supercooled glycerol: Evidence from wide field single molecule imaging”, *J. Chem. Phys.* **131** (2009).
- [60] B. G. Demirjian, G. Dosseh, A. Chauty, M.-L. Ferrer, D. Morineau, C. Lawrence, K. Takeda, D. Kivelson and S. Brown, “Metastable solid phase at the crystalline-amorphous border: the glacial phase of triphenyl phosphite”, *J. Phys. Chem. B* **105** (2001) 2107–2116.
- [61] H. Tanaka, R. Kurita and H. Mataka, “Liquid-liquid transition in the molecular liquid triphenyl phosphite”, *Phys. Rev. Lett.* **92** (2004) 025 701.
- [62] B. Bolshakov and A. Dzhonson, “On the number of amorphous phases in n-butanol: Kinetics of free radicals oxidation by oxygen in frozen n-butanol”, *J. Non-Cryst. Solids.* **351** (2005) 444–454.
- [63] V. M. Syutkin, V. L. Vyazovkin, V. V. Korolev and S. Y. Grebenkin, “Length and time scales of structural heterogeneities in deeply supercooled propylene carbonate”, *Phys. Rev. Lett.* **109** (2012).
- [64] M. Saito, S. Kitao, Y. Kobayashi, M. Kurokuzu, Y. Yoda and M. Seto, “Slow processes in supercooled o-Terphenyl: Relaxation and decoupling”, *Phys. Rev. Lett.* **109** (2012).
- [65] R. Zondervan, T. Xia, H. van der Meer, C. Storm, F. Kulzer, W. van Saarloos and M. Orrit, “Soft glassy rheology of supercooled molecular liquids”, *Proc. Natl. Acad. Sci. USA* **105** (2008) 4993–4998.
- [66] M. E. Mobius, T. Xia, W. van Saarloos, M. Orrit and M. van Hecke, “Aging and solidification of supercooled glycerol”, *J. Phys. Chem. B* **114** (2010) 7439–7444.
- [67] S. Das, “Mode-coupling theory and the glass transition in supercooled liquids”, *Rev. Mod. Phys.* **76** (2004) 785–851.
- [68] D. R. Reichman and P. Charbonneau, “Mode-coupling theory”, *J. Stat. Mech. Theor. Exp.* **2005** (2005) P05 013.
- [69] H. Chen, L. Shao, Q. Li and J. Wang, “Gold nanorods and their plasmonic properties”, *Chem. Soc. Rev.* **42** (2013) 2679–2724.
- [70] M. Yorulmaz, S. Khatua, P. Zijlstra, A. Gaiduk and M. Orrit, “Luminescence quantum yield of single gold nanorods”, *Nano Lett.* **12** (2012) 4385–4391.

References

- [71] T. Ming, H. Chen, R. Jiang, Q. Li and J. Wang, "Plasmon-controlled fluorescence: Beyond the intensity enhancement", *J. Phys. Chem. Lett.* **3** (2012) 191–202.
- [72] A. A. Kinkhabwala, Z. Yu, S. Fan and W. E. Moerner, "Fluorescence correlation spectroscopy at high concentrations using gold bowtie nanoantennas", *Chem. Phys.* **406** (2012) 3–8.
- [73] H. Yuan, S. Khatua, P. Zijlstra, M. Yorulmaz and M. Orrit, "Thousand-fold enhancement of single-molecule fluorescence near a single gold nanorod", *Angew. Chem. Int. Ed.* **52** (2013) 1217–1221.
- [74] J. D. Stevenson and P. G. Wolynes, "The ultimate fate of supercooled liquids", *J. Phys. Chem. A* **115** (2011) 3713–3719.
- [75] T. Ha, T. Enderle, D. F. Ogletree, D. S. Chemla, P. R. Selvin and S. Weiss, "Probing the interaction between two single molecules: fluorescence resonance energy transfer between a single donor and a single acceptor.", *Proc. Natl. Acad. Sci. U. S. A.* **93** (1996) 6264–8.
- [76] S. Hohng, C. Joo and T. Ha, "Single-molecule three-color FRET", *Biophys. J.* **87** (2004) 1328–1337.
- [77] X. Zhuang, L. E. Bartley, H. P. Babcock, R. Russell, T. Ha, D. Herschlag and S. Chu, "A single-molecule study of rna catalysis and folding.", *Science* **288** (2000) 2048–51.
- [78] N. K. Lee, A. N. Kapanidis, H. R. Koh, Y. Korlann, S. O. Ho, Y. Kim, N. Gassman, S. K. Kim and S. Weiss, "Three-color alternating-laser excitation of single molecules: monitoring multiple interactions and distances", *Biophys. J.* **92** (2007) 303–312.
- [79] B. Schuler, E. A. Lipman, P. J. Steinbach, M. Kumke and W. A. Eaton, "Polyproline and the "spectroscopic ruler" revisited with single-molecule fluorescence.", *Proc. Natl. Acad. Sci. U. S. A.* **102** (2005) 2754–9.
- [80] A. K. Wozniak, G. F. Schröder, H. Grubmüller, C. A. Seidel and F. Oesterhelt, "Single-molecule FRET measures bends and kinks in DNA.", *Proc. Natl. Acad. Sci. U. S. A.* **105** (2008) 18337–18342.
- [81] D. Nettels, S. Mueller-Spaeth, F. Kuester, H. Hofmann, D. Haenni, S. Rueegger, L. Reymond, A. Hoffmann, J. Kubelka, B. Heinz, K. Gast, R. B. Best and B. Schuler, "Single-molecule spectroscopy of the temperature-induced collapse of unfolded proteins", *Proc. Natl. Acad. Sci. U. S. A.* **106** (2009) 20740–20745.
- [82] A. Deniz, M. Dahan, J. Grunwell, T. Ha, A. Faulhaber, D. Chemla, S. Weiss and P. Schultz, "Single-pair fluorescence resonance energy transfer on freely diffusing molecules: Observation of Forster distance dependence and subpopulations", *Proc. Natl. Acad. Sci. U. S. A.* **96** (1999) 3670–3675.

-
- [83] R. E. Dale, J. Eisinger and W. E. Blumberg, “The orientational freedom of molecular probes. the orientation factor in intramolecular energy transfer.”, *Biophys. J.* **26** (1979) 161–93.
- [84] J. Eisinger, B. Feuer and A. A. Lamola, “Intramolecular singlet excitation transfer. Applications to polypeptides.”, *Biochemistry* **8** (1969) 3908–15.
- [85] K. A. Dill, S. B. Ozkan, T. R. Weikl, J. D. Chodera and V. A. Voelz, “The protein folding problem: when will it be solved?”, *Curr. Opin. Struc. Biol.* **17** (2007) 342–346.
- [86] L. Stryer and R. P. Haugland, “Energy transfer: a spectroscopic ruler.”, *Proc. Natl. Acad. Sci. U. S. A.* **58** (1967) 719–26.
- [87] K. Schröter and E. Donth, “Comparison of shear response with other properties at the dynamic glass transition of different glassformers”, *J. Non-Cryst. Solids* **307-310** (2002) 270–280.
- [88] J. L. Dashnau, N. V. Nucci, K. A. Sharp and J. M. Vanderkooi, “Hydrogen bonding and the cryoprotective properties of glycerol/water mixtures.”, *J. Phys. Chem. B* **110** (2006) 13 670–13 677.
- [89] A. M. Klibanov, “Improving enzymes by using them in organic solvents”, *Nature* **409** (2001) 241–246.
- [90] Timasheff, Serge N., “Solvent stabilization of protein structure”, in Shirley,, Bret .. (ed.), “Protein Stability and Folding”, vol. 40 of *Methods in Molecular Biology*, Humana Press (1995), pp. 253–269.
- [91] M. Auton and D. W. Bolen, “Additive transfer free energies of the peptide backbone unit that are independent of the model compound and the choice of concentration scale†”, *Biochemistry* **43** (2004) 1329–1342.
- [92] Eker, F., Griebenow, K. and Schweitzer-Stenner, R., “Stable conformations of tripeptides in aqueous solution studied by UV circular dichroism spectroscopy”, *J. Am. Chem. Soc.* **125** (2003) 8178–8185.
- [93] K. Schroter and E. Donth, “Viscosity and shear response at the dynamic glass transition of glycerol”, *J. Chem. Phys.* **113** (2000) 9101–9108.
- [94] V. Ivanov, M. Li and K. Mizuuchi, “Impact of emission anisotropy on fluorescence spectroscopy and FRET distance measurements”, *Biophys. J.* **97** (2009) 922–929.
- [95] F. Ye, M. M. Collinson and D. A. Higgins, “Molecular orientation and its influence on autocorrelation amplitudes in single-molecule imaging experiments”, *Anal. Chem.* **79** (2007) 6465–6472.
- [96] Z. Sikorski and L. M. Davis, “Engineering the collected field for single-molecule orientation determination”, *Opt. Express* **16** (2008) 3660–3673.

References

- [97] R. Zondervan, F. Kulzer, S. Orlinskii and M. Orrit, “Photoblinking of rhodamine 6G in poly(vinyl alcohol): Radical dark state formed through the triplet”, *J. Phys. Chem. A* **107** (2003) 6770–6776.
- [98] R. Zondervan, F. Kulzer, M. Kol’chenko and M. Orrit, “Photobleaching of rhodamine 6G in poly(vinyl alcohol) at the ensemble and single-molecule levels”, *J. Phys. Chem. A* **108** (2004) 1657–1665.
- [99] L. P. Watkins, H. Chang and H. Yang, “Quantitative single-molecule conformational distributions: A case study with poly-(l-proline)”, *J. Phys. Chem. A* **110** (2006) 5191–5203.
- [100] R. B. Best, K. A. Merchant, I. V. Gopich, B. Schuler, A. Bax and W. A. Eaton, “Effect of flexibility and cis residues in single-molecule FRET studies of polyproline”, *Proc. Natl. Acad. Sci. U. S. A.* **104** (2007) 18 964–18 969.
- [101] A. Paciaroni, S. Cinelli and G. Onori, “Effect of the environment on the protein dynamical transition: a neutron scattering study”, *Biophys. J.* **83** (2002) 1157–1164.
- [102] E. Cornicchi, S. Cinelli, F. Natali, G. Onori and A. Paciaroni, “Elastic neutron scattering study of proton dynamics in glycerol”, *Phys. B. Condens. Matter.* **350** (2004) E951–E954.
- [103] C. Chen, W. Z. Li, Y. C. Song and J. Yang, “Hydrogen bonding analysis of glycerol aqueous solutions: A molecular dynamics simulation study”, *J. Mol. Liq.* **146** (2009) 23 – 28.
- [104] A. Ansari, C. Jones, E. Henry, J. Hofrichter and W. Eaton, “The role of solvent viscosity in the dynamics of protein conformational-changes”, *Science* **256** (1992) 1796–1798.
- [105] R. Chelli, P. Procacci, G. Cardini and S. Califano, “Glycerol condensed phases part II. A molecular dynamics study of the conformational structure and hydrogen bonding”, *Phys. Chem. Chem. Phys.* **1** (1999) 879–885.
- [106] M. Gonnelli and G. Strambini, “Glycerol effects on protein flexibility: a tryptophan phosphorescence study”, *Biophys. J.* **65** (1993) 131–137.
- [107] S. A. McKinney, C. Joo and T. Ha, “Analysis of single-molecule FRET trajectories using hidden Markov modeling”, *Biophys. J.* **91** (2006) 1941–1951.
- [108] B. Schuler, E. Lipman and W. Eaton, “Probing the free-energy surface for protein folding with single-molecule fluorescence spectroscopy”, *Nature* **419** (2002) 743–747.
- [109] F. Hillger, D. Haenni, D. Nettels, S. Geister, M. Grandin, M. Textor and B. Schuler, “Probing protein-chaperone interactions with single-molecule fluorescence spectroscopy”, *Angew. Chem. Int. Ed.* **47** (2008) 6184–6188.

- [110] D. Nettels, S. Mueller-Spaeth, F. Kuester, H. Hofmann, D. Haenni, S. Rueegger, L. Reymond, A. Hoffmann, J. Kubelka, B. Heinz, K. Gast, R. B. Best and B. Schuler, “Single-molecule spectroscopy of the temperature-induced collapse of unfolded proteins”, *Proc. Natl. Acad. Sci. U. S. A.* **106** (2009) 20 740–20 745.
- [111] W. J. A. Koopmans, R. Buning, T. Schmidt and J. van Noort, “spFRET using alternating excitation and FCS reveals progressive DNA unwrapping in nucleosomes”, *Biophys. J.* **97** (2009) 195–204.
- [112] A. Gansen, A. Valeri, F. Hauger, S. Felekyan, S. Kalinin, K. Toth, J. Langowski and C. A. M. Seidel, “Nucleosome disassembly intermediates characterized by single-molecule FRET”, *Proc. Natl. Acad. Sci. U. S. A.* **106** (2009) 15 308–15 313.
- [113] J. Lakowicz, I. Gryczynski, G. Piszczek, L. Tolosa, R. Nair, M. Johnson and K. Nowaczyk, “Microsecond dynamics of biological macromolecules”, in “Energetics of biological macromolecules, pt C”, vol. 323 of *Methods Enzymol.* (2000), pp. 473–509.
- [114] L. A. Campos, J. Liu, X. Wang, R. Ramanathan, D. S. English and V. Munoz, “A photoprotection strategy for microsecond-resolution single-molecule fluorescence spectroscopy”, *Nat. Methods* **8** (2011) 143–U63.
- [115] H. Yuan, T. Xia, B. Schuler and M. Orrit, “Temperature-cycle single-molecule FRET microscopy on polyprolines”, *Phys. Chem. Chem. Phys.* **13** (2011) 1762–1769.
- [116] S. Doose, H. Neuweiler, H. Barsch and M. Sauer, “Probing polyproline structure and dynamics by photoinduced electron transfer provides evidence for deviations from a regular polyproline type II helix”, *Proc. Natl. Acad. Sci. U. S. A.* **104** (2007) 17 400–17 405.
- [117] S. Sidebert, S. Kalinin, N. Hien, A. Kienzler, L. Clima, W. Bannwarth, B. Appel, S. Muller and C. A. M. Seidel, “Accurate distance determination of nucleic acids via Förster resonance energy transfer: Implications of dye linker length and rigidity”, *J. Am. Chem. Soc.* **133** (2011) 2463–2480.
- [118] M. Hoeffling, N. Lima, D. Haenni, C. A. M. Seidel, B. Schuler and H. Grubmuller, “Structural heterogeneity and quantitative FRET efficiency distributions of polyprolines through a hybrid atomistic simulation and Monte Carlo approach”, *PLOS One* **6** (2011) 19.
- [119] D. Badali and C. C. Gradinaru, “The effect of Brownian motion of fluorescent probes on measuring nanoscale distances by Förster resonance energy transfer”, *J. Chem. Phys.* **134** (2011) 11.
- [120] A. Iqbal, S. Arslan, B. Okumus, T. J. Wilson, G. Giraud, D. G. Norman, T. Ha and D. M. J. Lilley, “Orientation dependence in fluorescent energy transfer between Cy3 and Cy5 terminally attached to double-stranded nucleic acids”, *Proc. Natl. Acad. Sci. U. S. A.* **105** (2008) 11 176–11 181.

- [121] J. Ouellet, S. Schorr, A. Iqbal, T. J. Wilson and D. M. J. Lilley, "Orientation of cyanine fluorophores terminally attached to dna via long, flexible tethers", *Biophys. J.* **101** (2011) 1148–1154.
- [122] W. Kugel, A. Muschielok and J. Michaelis, "Bayesian-inference-based fluorescence correlation spectroscopy and single-molecule burst analysis reveal the influence of dye selection on DNA hairpin dynamics", *ChemPhysChem* **13** (2012) 1013–1022.
- [123] H. S. Chung, J. M. Louis and W. A. Eaton, "Distinguishing between protein dynamics and dye photophysics in single-molecule FRET experiments", *Biophys. J.* **98** (2010) 696–706.
- [124] N. Di Fiori and A. Meller, "The effect of dye-dye interactions on the spatial resolution of single-molecule FRET measurements in nucleic acids", *Biophys. J.* **98** (2010) 2265–2272.
- [125] S. Kalinin, E. Sisamakos, S. W. Magennis, S. Felekyan and C. A. M. Seidel, "On the origin of broadening of single-molecule FRET efficiency distributions beyond shot noise limits", *J. Phys. Chem. B* **114** (2010) 6197–6206.
- [126] M. Antonik, S. Felekyan, A. Gaiduk and C. A. M. Seidel, "Separating structural heterogeneities from stochastic variations in fluorescence resonance energy transfer distributions via photon distribution analysis", *J. Phys. Chem. B* **110** (2006) 6970–6978.
- [127] E. Nir, X. Michalet, K. M. Hamadani, T. A. Laurence, D. Neuhauser, Y. Kovchegov and S. Weiss, "Shot-noise limited single-molecule FRET histograms: Comparison between theory and experiments", *J. Phys. Chem. B* **110** (2006) 22 103–22 124.
- [128] S. S. Vogel, T. A. Nguyen, B. W. van der Meer and P. S. Blank, "The impact of heterogeneity and dark acceptor states on fret: Implications for using fluorescent protein donors and acceptors", *PLoS ONE* **7** (2012) e49593.
- [129] R. Brune, S. Doose and M. Sauer, "Analyzing the influence of contact-induced quenching processes on förster resonance energy transfer", (2007) 66 331M–66 331M–11.
- [130] S. M. Mansfield and G. S. Kino, "Solid immersion microscope", *Appl. Phys. Lett.* **57** (1990) 2615–2616.
- [131] K. Koyama, M. Yoshita, M. Baba, T. Suemoto and H. Akiyama, "High collection efficiency in fluorescence microscopy with a solid immersion lens", *Appl. Phys. Lett.* **75** (1999) 1667–1669.
- [132] T. D. Milster, J. S. Jo and K. Hirota, "Roles of propagating and evanescent waves in solid immersion lens systems", *Appl. Opt.* **38** (1999) 5046–5057.
- [133] K. Karrai, X. Lorenz and L. Novotny, "Enhanced reflectivity contrast in confocal solid immersion lens microscopy", *Appl. Phys. Lett.* **77** (2000) 3459–3461.

-
- [134] S. Moehl, H. Zhao, B. D. Don, S. Wachter and H. Kalt, “Solid immersion lens-enhanced nano-photoluminescence: Principle and applications”, *J. Appl. Phys.* **93** (2003) 6265–6272.
- [135] T. Schröder, F. Gädeke, M. J. Banholzer and O. Benson, “Ultrabright and efficient single-photon generation based on nitrogen-vacancy centres in nanodiamonds on a solid immersion lens”, *New J. Phys.* **13** (2011) 055 017.
- [136] M. Celebrano, R. Lettow, P. Kukura, M. Agio, A. Renn, S. Götzinger and V. Sandoghdar, “Efficient coupling of single photons to single plasmons”, *Opt. Express* **18** (2010) 13 829–13 835.
- [137] Y. L. A. Rezus, S. G. Walt, R. Lettow, A. Renn, G. Zumofen, S. Götzinger and V. Sandoghdar, “Single-photon spectroscopy of a single molecule”, *Phys. Rev. Lett.* **108** (2012) 093 601.
- [138] S. Fujiyoshi, M. Fujiwara, C. Kim, M. Matsushita, A. M. van Oijen and J. Schmidt, “Single-component reflecting objective for low-temperature spectroscopy in the entire visible region”, *Appl. Phys. Lett.* **91** (2007) 051 125.
- [139] S. Fujiyoshi, M. Fujiwara and M. Matsushita, “Visible fluorescence spectroscopy of single proteins at liquid-helium temperature”, *Phys. Rev. Lett.* **100** (2008) 168 101.
- [140] J. P. Torella, S. J. Holden, Y. Santoso, J. Hohlbein and A. N. Kapanidis, “Identifying molecular dynamics in single-molecule FRET experiments with burst variance analysis”, *Biophys. J.* **100** (2011) 1568–1577.
- [141] T. Tomov, R. Tsukanov, R. Masoud, M. Liber, N. Plavner and E. Nir, “Disentangling subpopulations in single-molecule FRET and ALEX experiments with photon distribution analysis”, *Biophys. J.* **102** (2012) 1163–1173.
- [142] M. I. Wallace, L. Ying, S. Balasubramanian and D. Klenerman, “FRET fluctuation spectroscopy: exploring the conformational dynamics of a DNA hairpin loop”, *J. Phys. Chem. B* **104** (2000) 11 551–11 555.
- [143] C. A. Angell, “Formation of glasses from liquids and biopolymers”, *Science* **267** (1995) 1924–1935.
- [144] F. Fujara, W. Petry, R. M. Diehl, W. Schnauss and H. Sillescu, “localized motion in supercooled glycerol as measured by h-2-nmr spin-lattice relaxation and incoherent neutron-scattering”, *Europhys. Lett.* **14** (1991) 563–568.
- [145] J. Wuttke, J. Hernandez, G. Li, G. Coddens, H. Z. Cummins, F. Fujara, W. Petry and H. Sillescu, “Neutron and light-scattering study of supercooled glycerol”, *Phys. Rev. Lett.* **72** (1994) 3052–3055.
- [146] A. Schob, F. Cichos, J. Schuster and C. von Borczyskowski, “Reorientation and translation of individual dye molecules in a polymer matrix”, *Eur. Polym. J.* **40** (2004) 1019–1026.

References

- [147] S. A. Mackowiak, L. M. Leone and L. J. Kaufman, “Probe dependence of spatially heterogeneous dynamics in supercooled glycerol as revealed by single molecule microscopy”, *Phys. Chem. Chem. Phys.* **13** (2011) 1786–1799.
- [148] T. Xia, L. T. Xiao and M. Orrit, “Micron-sized structure in a thin glycerol film revealed by fluorescent probes”, *J. Phys. Chem. B* **113** (2009) 15 724–15 729.
- [149] A. Ha, I. Cohen, X. L. Zhao, M. Lee and D. Kivelson, “Supercooled liquids and polyamorphism”, *J. Phys. Chem.* **100** (1996) 1–4.
- [150] I. M. Shmytko, R. J. Jiménez-Riobóo, M. Hassaine and M. A. Ramos, “Structural and thermodynamic studies of n-butanol”, *J. Phys.: Condens. Matter.* **22** (2010) 195 102.
- [151] B. E. Schwickert, S. R. Kline, H. Zimmermann, K. M. Lantzky and J. L. Yarger, “Early stages of glacial clustering in supercooled triphenyl phosphite”, *Phys. Rev. B* **64** (2001) 6.
- [152] S. A. Kivelson, X. Zhao, D. Kivelson, T. M. Fischer and C. M. Knobler, “Frustration-limited clusters in liquids”, *J. Chem. Phys.* **101** (1994) 2391–2397.
- [153] C. Alba-Simionesco and G. Tarjus, “Experimental evidence of mesoscopic order in the apparently amorphous glacial phase of the fragile glass former triphenylphosphite”, *Europhys. Lett.* **52** (2000) 297–303.
- [154] G. Tarjus, S. A. Kivelson, Z. Nussinov and P. Viot, “The frustration-based approach of supercooled liquids and the glass transition: a review and critical assessment”, *J. Phys.: Condens. Matter.* **17** (2005) R1143.
- [155] A. Hedoux, Y. Guinet, P. Derollez, O. Hernandez, R. Lefort and M. Descamps, “A contribution to the understanding of the polyamorphism situation in triphenyl phosphite”, *Phys. Chem. Chem. Phys.* **6** (2004) 3192–3199.
- [156] A. Hedoux, Y. Guinet, P. Derollez, O. Hernandez, L. Paccou and M. Descamps, “Micro-structural investigations in the glacial state of triphenyl phosphite”, *J. Non-Cryst. Solids.* **352** (2006) 4994–5000.
- [157] Q. Mei, P. Ghalsasi, C. J. Benmore and J. L. Yarger, “The local structure of triphenyl phosphite studied using spallation neutron and high-energy X-ray diffraction”, *J. Phys. Chem. B* **108** (2004) 20 076–20 082.
- [158] V. Lubchenko and P. G. Wolynes, “Theory of structural glasses and supercooled liquids”, *Annu. Rev. Phys. Chem.* **58** (2007) 235–266.
- [159] K. L. Ngai, “Dynamic and thermodynamic properties of glass-forming substances”, *J. Non-Cryst. Solids.* **275** (2000) 7–51.
- [160] F. J. Bermejo, A. Criado, A. d. d. deAndres, E. Enciso and H. Schober, “Microscopic dynamics of glycerol in its crystalline and glassy states”, *Phys. Rev. B* **53** (1996) 5259–5267.

-
- [161] I. B. Rabinovich, V. I. Murzin and L. S. Zhilkin, “The isotropic effect in the viscosity of deuteroglycerine and ethylenedeglycerol”, *Zhur. Fiz. Khim.* **34** (1960) 1973.
- [162] A. L. Patterson, “The Scherrer formula for X-ray particle size determination”, *Phys. Rev.* **56** (1939) 978–982.
- [163] J. D. Stevenson, J. Schmalian and P. G. Wolynes, “The shapes of cooperatively rearranging regions in glass-forming liquids”, *Nat. Phys.* **2** (2006) 268–274.
- [164] S. A. Reinsberg, X. H. Qiu, M. Wilhelm, H. W. Spiess and M. D. Ediger, “Length scale of dynamic heterogeneity in supercooled glycerol near T_g ”, *J. Chem. Phys.* **114** (2001) 7299–7302.
- [165] M. D. Ediger and P. Harrowell, “Perspective: Supercooled liquids and glasses”, *J. Chem. Phys.* **137** (2012) 080 901.
- [166] U. Tracht, M. Wilhelm, A. Heuer, H. Feng, K. Schmidt-Rohr and H. Spiess, “Length scale of dynamic heterogeneities at the glass transition determined by multidimensional nuclear magnetic resonance”, *Phys. Rev. Lett.* **81** (1998) 2727–2730.
- [167] F. Qi, T. El Goresy, R. Bohmer, A. Doss, G. Diezemann, G. Hinze, H. Sillescu, T. Blochowicz, C. Gainaru, E. Rossler and H. Zimmermann, “Nuclear magnetic resonance and dielectric spectroscopy of a simple supercooled liquid: 2-methyl tetrahydrofuran”, *J. Chem. Phys.* **118** (2003) 7431–7438.
- [168] R. Böhmer, G. Hinze, G. Diezemann, B. Geil and H. Sillescu, “Dynamic heterogeneity in supercooled ortho-terphenyl studied by multidimensional deuteron NMR”, *Europhys. Lett.* **36** (1996) 55.
- [169] G. Diezemann, R. Böhmer, G. Hinze and H. Sillescu, “Reorientational dynamics in simple supercooled liquids”, *J. Non-Cryst. Solids* **235-237** (1998) 121–127.
- [170] U. Schneider, P. Lunkenheimer, R. Brand and A. Loidl, “Dielectric and far-infrared spectroscopy of glycerol”, *J. Non-Cryst. Solids* **235-237** (1998) 173–179.
- [171] A. Patkowski, T. Thurn-Albrecht, E. Banachowicz, W. Steffen, P. Bösecke, T. Narayanan and E. W. Fischer, “Long-range density fluctuations in orthoterphenyl as studied by means of ultrasmall-angle x-ray scattering”, *Phys. Rev. E* **61** (2000) 6909–6913.
- [172] A. Patkowski, E. W. Fischer, W. Steffen, H. Gläser, M. Baumann, T. Ruths and G. Meier, “Unusual features of long-range density fluctuations in glass-forming organic liquids: A Rayleigh and Rayleigh-Brillouin light scattering study”, *Phys. Rev. E* **63** (2001) 061 503.
- [173] A. S. Bakai and E. W. Fischer, “Nature of long-range correlations of density fluctuations in glass-forming liquids”, *J. Chem. Phys.* **120** (2004) 5235–5252.

- [174] P. Zijlstra and M. Orrit, “Single metal nanoparticles: optical detection, spectroscopy and applications”, *Rep. Prog. Phys.* **74** (2011) 106 401.
- [175] J. Rodríguez-Fernández, J. Pérez-Juste, L. M. Liz-Marzán and P. R. Lang, “Dynamic light scattering of short au rods with low aspect ratios”, *J. Phys. Chem. C* **111** (2007) 5020–5025.
- [176] N. K. Reddy, J. Pérez-Juste, I. Pastoriza-Santos, P. R. Lang, J. K. G. Dhont, L. M. Liz-Marzán and J. Vermant, “Flow dichroism as a reliable method to measure the hydrodynamic aspect ratio of gold nanoparticles”, *ACS Nano* **5** (2011) 4935–4944.
- [177] W.-S. Chang, J. W. Ha, L. S. Slaughter and S. Link, “Plasmonic nanorod absorbers as orientation sensors”, *Proc. Natl. Acad. Sci. USA* **107** (2010) 2781–2786.
- [178] A. Tcherniak, S. Dominguez-Medina, W.-S. Chang, P. Swanglap, L. S. Slaughter, C. F. Landes and S. Link, “One-photon plasmon luminescence and its application to correlation spectroscopy as a probe for rotational and translational dynamics of gold nanorods”, *J. Phys. Chem. C* **115** (2011) 15 938–15 949.
- [179] H. Wang, T. B. Huff, D. A. Zweifel, W. He, P. S. Low, A. Wei and J.-X. Cheng, “In vitro and in vivo two-photon luminescence imaging of single gold nanorods”, *Proc. Natl. Acad. Sci. USA* **102** (2005) 15 752–15 756.
- [180] B. Nikoobakht and M. A. El-Sayed, “Preparation and growth mechanism of gold nanorods (NRs) using seed-mediated growth method”, *Chem. Mater.* **15** (2003) 1957–1962.
- [181] H.-F. Yuan, T. Xia, M. Plazanet, B. Demé and M. Orrit, “Communication: Crystallite nucleation in supercooled glycerol near the glass transition”, *J. Chem. Phys.* **136** (2012) 041 102.
- [182] C.-Y. Lu and D. A. Vanden Bout, “Effect of finite trajectory length on the correlation function analysis of single molecule data”, *J. Chem. Phys.* **125** (2006).
- [183] H. Petrova, J. Perez Juste, I. Pastoriza-Santos, G. V. Hartland, L. M. Liz-Marzán and P. Mulvaney, “On the temperature stability of gold nanorods: comparison between thermal and ultrafast laser-induced heating”, *Phys. Chem. Chem. Phys.* **8** (2006) 814–821.
- [184] H. Ma, P. M. Bendix and L. B. Oddershede, “Large-scale orientation dependent heating from a single irradiated gold nanorod”, *Nano Lett.* **12** (2012) 3954–3960.
- [185] Z. J. Coppens, W. Li, D. G. Walker and J. G. Valentine, “Probing and controlling photothermal heat generation in plasmonic nanostructures”, *Nano Lett.* **0** (0) null.
- [186] P. V. Ruijgrok, N. R. Verhart, P. Zijlstra, A. L. Tchebotareva and M. Orrit, “Brownian fluctuations and heating of an optically aligned gold nanorod”, *Phys. Rev. Lett.* **107** (2011) 037 401.

-
- [187] D. Rings, D. Chakraborty and K. Kroy, “Rotational hot Brownian motion”, *New J. Phys.* **14** (2012) 053 012.
- [188] X. Qiu and M. D. Ediger, “Length scale of dynamic heterogeneity in supercooled d-sorbitol: comparison to model predictions”, *J. Phys. Chem. B* **107** (2003) 459–464.
- [189] R. Kurita and H. Tanaka, “Critical-like phenomena associated with liquid-liquid transition in a molecular liquid”, *Science* **306** (2004) 845–848.
- [190] M. P. Backlund, M. D. Lew, A. S. Backer, S. J. Sahl, G. Grover, A. Agrawal, R. Piestun and W. E. Moerner, “Simultaneous, accurate measurement of the 3D position and orientation of single molecules”, *Proc. Natl. Acad. Sci. USA* **109** (2012) 19 087–19 092.
- [191] J. Foelling, M. Bossi, H. Bock, R. Medda, C. A. Wurm, B. Hein, S. Jakobs, C. Eggeling and S. W. Hell, “Fluorescence nanoscopy by ground-state depletion and single-molecule return”, *Nat. Methods* **5** (2008) 943–945.
- [192] M. Heilemann, P. Dedecker, J. Hofkens and M. Sauer, “Photoswitches: Key molecules for subdiffraction-resolution fluorescence imaging and molecular quantification”, *Laser Photon. Rev.* **3** (2009) 180–202.
- [193] L. M. Leone and L. J. Kaufman, “Single molecule probe reports of dynamic heterogeneity in supercooled ortho-terphenyl”, *J. Chem. Phys.* **138** (2013) 12A524.
- [194] K. Velonia, O. Flomenbom, D. Loos, S. Masuo, M. Cotlet, Y. Engelborghs, J. Hofkens, A. E. Rowan, J. Klafter, R. J. M. Nolte and F. C. de Schryver, “Single-enzyme kinetics of CALB-catalyzed hydrolysis”, *Angew. Chem.* **117** (2005) 566–570.
- [195] B. Huang, M. Bates and X. Zhuang, “Super-resolution fluorescence microscopy”, *Annu. Rev. Biochem.* **78** (2009) 993–1016.
- [196] M. Lakadamyali, M. Rust and X. Zhuang, “Ligands for clathrin-mediated endocytosis are differentially sorted into distinct populations of early endosomes”, *Cell* **124** (2006) 997–1009.
- [197] P. Zijlstra, P. M. R. Paulo and M. Orrit, “Optical detection of single non-absorbing molecules using the surface plasmon resonance of a gold nanorod”, *Nat. Nanotechnol.* **7** (2012) 379–382.
- [198] M. Liu, P. Guyot-Sionnest, T.-W. Lee and S. K. Gray, “Optical properties of rodlike and bipyramidal gold nanoparticles from three-dimensional computations”, *Phys. Rev. B* **76** (2007) 235 428.
- [199] O. L. Muskens, V. Giannini, J. A. Sánchez-Gil and J. Gómez Rivas, “Strong enhancement of the radiative decay rate of emitters by single plasmonic nanoantennas”, *Nano Lett.* **7** (2007) 2871–2875.

References

- [200] A. F. Koenderink, “On the use of Purcell factors for plasmon antennas”, *Opt. Lett.* **35** (2010) 4208–4210.
- [201] W. P. Ambrose, P. M. Goodwin, R. A. Keller and J. C. Martin, “Alterations of single molecule fluorescence lifetimes in near-field optical microscopy”, *Science* **265** (1994) 364–367.
- [202] S. Kühn, U. Håkanson, L. Rogobete and V. Sandoghdar, “Enhancement of single-molecule fluorescence using a gold nanoparticle as an optical nanoantenna”, *Phys. Rev. Lett.* **97** (2006) 017402.
- [203] P. Anger, P. Bharadwaj and L. Novotny, “Enhancement and quenching of single-molecule fluorescence”, *Phys. Rev. Lett.* **96** (2006) 113002.
- [204] A. G. Curto, G. Volpe, T. H. Taminiau, M. P. Kreuzer, R. Quidant and N. F. van Hulst, “Unidirectional emission of a quantum dot coupled to a nanoantenna”, *Science* **329** (2010) 930–933.
- [205] J. N. Farahani, D. W. Pohl, H.-J. Eisler and B. Hecht, “Single quantum dot coupled to a scanning optical antenna: a tunable superemitter”, *Phys. Rev. Lett.* **95** (2005) 017402.
- [206] A. Kinkhabwala, Z. Yu, S. Fan, Y. Avlasevich, K. Muellen and W. E. Moerner, “Large single-molecule fluorescence enhancements produced by a bowtie nanoantenna”, *Nature Photon.* **3** (2009) 654–657.
- [207] P. Zijlstra, A. L. Tchebotareva, J. W. M. Chon, M. Gu and M. Orrit, “Acoustic oscillations and elastic moduli of single gold nanorods”, *Nano Lett.* **8** (2008) 3493–3497.
- [208] G. Lu, T. Zhang, W. Li, L. Hou, J. Liu and Q. Gong, “Single-molecule spontaneous emission in the vicinity of an individual gold nanorod”, *J. Phys. Chem. C* **115** (2011) 15822–15828.
- [209] A. Mohammadi, V. Sandoghdar and M. Agio, “Gold nanorods and nanospheroids for enhancing spontaneous emission”, *New J. Phys.* **10** (2008) 105015.
- [210] Y. Fu, J. Zhang and J. R. Lakowicz, “Plasmon-enhanced fluorescence from single fluorophores end-linked to gold nanorods”, *J. Am. Chem. Soc.* **132** (2010) 5540–5541.
- [211] P. Bharadwaj, R. Beams and L. Novotny, “Nanoscale spectroscopy with optical antennas”, *Chem. Sci.* **2** (2011) 136–140.
- [212] L. A. Brey, G. B. Schuster and H. G. Drickamer, “High pressure studies of the effect of viscosity on fluorescence efficiency in crystal violet and auramine O”, *J. Chem. Phys.* **67** (1977) 2648–2650.
- [213] Y. Fang, W.-S. Chang, B. Willingham, P. Swanglap, S. Dominguez-Medina and S. Link, “Plasmon emission quantum yield of single gold nanorods as a function of aspect ratio”, *ACS Nano* **6** (2012) 7177–7184.

- [214] E. Haustein and P. Schwille, “Fluorescence correlation spectroscopy: Novel variations of an established technique”, *Annu. Rev. Biophys. Biomol. Struct.* **36** (2007) 151–169.
- [215] P. Schwille, J. Bieschke and F. Oehlenschläger, “Kinetic investigations by fluorescence correlation spectroscopy: The analytical and diagnostic potential of diffusion studies”, *Biophys. Chem.* **66** (1997) 211–228.

References
