

AD-A255 333

(2)

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE



d  
0188

REPORT DOCUMENTATION PAGE

1a REPORT SECURITY CLASSIFICATION <b>UNCLASSIFIED</b>		1b RESTRICTIVE MARKINGS	
2a SECURITY CLASSIFICATION AUTHORITY		3 DISTRIBUTION/AVAILABILITY OF REPORT This document has been approved for public release and sale; its distribution is unlimited.	
2b DECLASSIFICATION/DOWNGRADING SCHEDULE		4 PERFORMING ORGANIZATION REPORT NUMBER(S)	
5. MONITORING ORGANIZATION REPORT NUMBER(S)		6a NAME OF PERFORMING ORGANIZATION IBM Research Division Almaden Research Center	
6b OFFICE SYMBOL (if applicable)		7a. NAME OF MONITORING ORGANIZATION Office of Naval Research	
6c ADDRESS (City, State, and ZIP Code) 650 Harry Road San Jose, CA 95120-6099		7b ADDRESS (City, State, and ZIP Code) Chemistry Division Code 1113 Arlington, VA 22217	
8a NAME OF FUNDING/SPONSORING ORGANIZATION Office of Naval Research		8b OFFICE SYMBOL (if applicable)	
9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER N00014-84-C-0708 and N00014-91-C-0166, 4131022		10. SOURCE OF FUNDING NUMBERS	
11. TITLE (Include Security Classification) Final Report		PROGRAM ELEMENT NO	PROJECT NO
12. PERSONAL AUTHOR(S) W. E. Moerner		TASK NO	WORK UNIT ACCESSION NO
13a. TYPE OF REPORT Final	13b. TIME COVERED FROM 1984 TO 1992	14. DATE OF REPORT (Year, Month, Day) 1992 Aug 31	
15. PAGE COUNT 6			
16. SUPPLEMENTARY NOTATION			
17. COSATI CODES		18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)	
FIELD	GROUP	SUB-GROUP	
19. ABSTRACT (Continue on reverse if necessary and identify by block number)			
<b>DTIC</b> <b>SEP 09 1992</b> <b>92-24759</b> <b>92 9 00 027</b> <b>7P8</b>			
20. DISTRIBUTION/AVAILABILITY OF ABSTRACT <input checked="" type="checkbox"/> UNCLASSIFIED/UNLIMITED <input type="checkbox"/> SAME AS RPT <input type="checkbox"/> DTIC USERS		21. ABSTRACT SECURITY CLASSIFICATION <b>UNCLASSIFIED</b>	
22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. W.E. Moerner		22b. TELEPHONE (Include Area Code) (408) 927-2426	22c. OFFICE SYMBOL

OFFICE OF NAVAL RESEARCH

FINAL REPORT

COVERING WORK PARTIALLY SUPPORTED DURING THE YEARS 1984-1992

for

Contracts N00014-84-C-0708 and N00014-91-C-0166

R&T Code 4131022

Photochemical and Photophysical Dynamics of  
Persistent Spectral Hole-Burning, Photorefractivity,  
and Single Molecular Absorbers in Condensed Matter

W. E. Moerner

IBM Research Division, Almaden Research Center  
650 Harry Road, San Jose, California 95120-6099

31 August 1992

Reproduction in whole, or in part, is permitted for any purpose of the United States  
Government.

\* This document has been approved for public release and sale; its distribution is un-  
limited.

During the period of this contract, the following milestones have been achieved: the first detailed studies of hole-burning bottlenecks for organic and inorganic systems; first use of high resolution ultrasonic modulation to detect photochemical holes; first observation of photochemical hole production in 100 ns; first complete analysis of coupling, reading-writing constraints for single-photon hole-burning materials leading to the need for photon-gating; first observation of photon-gated hole-burning in an organic system; first observation of photon-gating via a donor-acceptor electron transfer mechanism, which allows fast (30 ns) hole formation in small focused laser spots; first observation of statistical fine structure in an inhomogeneously broadened spectral line; first optical detection and spectroscopy of a single impurity molecule in a solid; first temperature dependence of photon-gating to 90 K; first intracavity second harmonic generation in an organic crystal; first phase-sensitive, time-resolved study of ballistic phonon propagation in a solid; first direct observation of spectral diffusion in a solid using a single-molecule probe; first observation of lifetime-limited linewidths, dephasing, and nonlinear saturation for a single molecule; first observation of hole-burning and spectral diffusion for a single molecule in a polymer; first observation of photoinduced reaction kinetics for a single molecule; first observation of photon antibunching for a single molecule in a solid; first observation of hole-burning and spectral diffusion for a single molecule in a polymer; first observation of photon antibunching for a single molecule in a solid; first observation of photorefractivity in a polymer; and first demonstration of two-beam coupling in a photorefractive polymer.

The following postdoctoral Visiting Scientists have been supported under this contract: Alan Huston, Howard W. H. Lee, Thomas P. Carter, Lothar Kador, Stephen Ducharme, W. Pat Ambrose, and Thomas Basché.

This work is summarized in the thirty-seven interim technical reports presented to the Office of Naval Research and in the following publications:

1. A. L. Huston and W. E. Moerner, "Detection of Persistent Spectral Holes Using Ultrasonic Modulation," *J. Opt. Soc. Am. B: Optical Physics* **1**, 349 (1984).
2. M. Romagnoli, W. E. Moerner, F. M. Schellenberg, M. D. Levenson, and G. C. Bjorklund, "Beyond the Bottleneck: Submicrosecond Hole-Burning in Phthalocyanine," *J. Opt. Soc. Am. B: Optical Physics* **1**, 341 (1984).
3. W. E. Moerner, M. Gehrtz, and A. L. Huston, "On the Measurement of Quantum Efficiencies for Persistent Spectral Hole-Burning," *J. Phys. Chem.* **88**, 6459 (1984).
4. W. E. Moerner, "A Challenge for Laser Spectroscopy of Solids: Frequency Domain Optical Storage," *Photonics Spectra* **19**, 59 (February 1985).
5. H. W. H. Lee, A. L. Huston, M. Gehrtz, and W. E. Moerner, "Photochemical Hole-Burning in a Protonated Phthalocyanine with GaAlAs Diode Lasers," *Chem. Phys. Lett.* **114**, 491 (1985).
6. W. E. Moerner and M. D. Levenson, "Can Single-Photon Processes Provide Useful Materials for Frequency Domain Optical Storage?," *J. Opt. Soc. Amer. B: Optical Physics* **2**, 915 (1985).

DTIC QUALITY INSPECTED 1

1st | Special  
A-1 |  
100%

Code

and/or

Special

7. W. E. Moerner, E. M. Schellenberg, G. C. Bjorklund, P. Kaipa, and F. Lüty, "High Efficiency Photochemical Hole-Burning for an Infrared Color Center," *Phys. Rev. B*, **32**, 1270 (1985).
8. M. Gehrtz, W. E. Moerner, and G. C. Bjorklund, "Shot-Noise Limited Detection in FM Spectroscopy by Optical Nulling of Residual Amplitude Modulation," IBM RJ#4678.
9. H. W. H. Lee, M. Gehrtz, E. Marinero, and W. E. Moerner, "Two-Color, Photon-Gated Spectral Hole-Burning in an Organic Material," *Chem. Phys. Lett.* **118**, 611 (1985).
10. W. E. Moerner, "Laser-Light-Induced Physical Processes in Optical Materials: Persistent Spectral Hole-Burning," *Proc. Soc. Photo-Opt. Instr. Engr.* **541**, 60 (1985).
11. W. E. Moerner, R. M. Macfarlane, and R. M. Shelby, "Physics/Optics News in 1985: Photon-Gated Spectral Hole-Burning," IBM RJ#4796, *Optics News* **11** (12), 9 (1985).
12. W. E. Moerner, "Molecular Electronics for Frequency Domain Optical Storage: Persistent Spectral Hole-Burning - A Review," *J. Molec. Elec.* **1**, 55 (1985).
13. W. E. Moerner, P. Pokrowsky, E. M. Schellenberg, and G. C. Bjorklund, "Spectral Hole Dynamics for R' Centers in LiF," *Phys. Rev. B*, **33**, 5702 (1986).
14. W. E. Moerner and A. L. Huston, "Phase-Sensitive Ultrasonic Modulation of Persistent Spectral Holes," *Appl. Phys. Lett.* **48**, 1181 (1986).
15. W. Lenth and W. E. Moerner, "Gated Spectral Hole-Burning for Frequency Domain Optical Recording," *Optics Commun.* **58**, 249 (1986).
16. W. E. Moerner, "Dynamical Hole-Burning Requirements for Frequency Domain Optical Storage," in *Unconventional Photoactive Solids*, Harvey Scher, editor, (Plenum, New York, 1988), pp. 41-51.
17. W. Lenth, R. M. Macfarlane, W. E. Moerner, E. M. Schellenberg, R. M. Shelby, and G. C. Bjorklund, "High-Density Frequency-Domain Optical Recording," *Proc. Soc. Photo-opt. Instrum. Engr.* **695**, 216 (1986).
18. A. J. Sievers and W. E. Moerner, "Persistent Infrared Spectral Hole-Burning for Impurity Vibrational Modes in Solids," Chapter 6 of *Persistent Spectral Hole-Burning: Science and Applications*, W. E. Moerner, editor, Topics in Current Physics Vol. 44 (Springer, Berlin, Heidelberg, 1988).
19. W. E. Moerner and A. L. Huston, "Phase-sensitive Detection of Persistent Spectral Holes Using Synchronous Ultrasonic Modulation Spectroscopy," *J. Opt. Soc. Am. B: Opt. Phys.* **3**, P210, (1986).

20. W. E. Moerner, T. P. Carter, and C. Bräuchle, "Fast Burning of Persistent Spectral Holes in Small Laser Spots Using Photon-Gated Materials," *Appl. Phys. Lett.* **50**, 430 (1987).
21. T. P. Carter, C. Bräuchle, V. Y. Lee, M. Manavi, and W. E. Moerner, "Photon-Gated Spectral Hole-Burning Via Donor-Acceptor Electron Transfer," *Optics Letters* **12**, 370 (1987).
22. T. P. Carter, C. Bräuchle, V. Y. Lee, M. Manavi, and W. E. Moerner, "Mechanism of Photon-Gated Persistent Spectral Hole-Burning in Metalloporphyrin/Halomethane Systems: Donor-Acceptor Electron Transfer," *J. Phys. Chem.* **91**, 3998 (1987).
23. W. E. Moerner, W. Lenth, and G. C. Bjorklund, "Frequency Domain Optical Storage and Other Applications of Persistent Spectral Hole-Burning," Chapter 7 of *Persistent Spectral Hole-Burning: Science and Applications*, W. E. Moerner, editor, Topics in Current Physics Vol. 44 (Springer, Berlin, Heidelberg, 1988)
24. W. E. Moerner, "Introduction to Persistent Spectral Hole-Burning: Science and Applications," Chapter 1 of *Persistent Spectral Hole-Burning: Science and Applications*, W. E. Moerner, editor, Topics in Current Physics Vol. 44 (Springer, Berlin, Heidelberg, 1988).
25. W. E. Moerner and T. P. Carter, "Statistical Fine Structure in Inhomogeneously Broadened Absorption Lines," *Phys. Rev. Lett.*, **59**, 2705 (1987).
26. W. E. Moerner and T. P. Carter, "Statistical Fine Structure in Inhomogeneously Broadened Absorption Lines in Solids," AIP Conference Proceedings **172**: Advances in Laser Science III, (AIP, New York, 1988), p. 419.
27. T. P. Carter, M. Manavi, and W. E. Moerner, "Statistical Fine Structure in the Inhomogeneously Broadened Electronic Origin of Pentacene in *p*-Terphenyl," *J. Chem. Phys.* **89**, 1768 (1988).
28. T. P. Carter, D. E. Horne, and W. E. Moerner, "Pseudo-Stark Effect and EM/Stark Double-Modulation Spectroscopy for the Detection of Statistical Fine Structure in Alexandrite," *Chem. Phys. Lett.* **151**, 102 (1988).
29. L. Kador, T. P. Carter, and W. E. Moerner, "EM-Stark Double-Modulation Spectroscopy for the Detection of Weak Spectral Features in Solids," IEEE Lasers and Electro-Optics Society Annual Meeting Proceedings, IEEE Cat. No. 88CH2683-1, pp. 246-248 (1988).
30. W. E. Moerner and L. Kador, "Optical Detection and Spectroscopy of Single Molecules in Solids," *Phys. Rev. Lett.* **62**, 2535 (1989).
31. L. Kador, D. E. Horne, and W. E. Moerner, "Optical Detection and Probing of Single Dopant Molecules of Pentacene in a *p*-Terphenyl Host Crystal by Means of Absorption Spectroscopy," Feature Article in *J. Phys. Chem.* **94**, 1237 (1990).

32. W. E. Moerner and L. Kador, "Finding a Single Molecule in a Haystack: Optical Detection and Spectroscopy of Single Absorbers in Solids," *Analytical Chemistry*, **61**, 1217A (1989).
33. W. E. Moerner, "Photon-Gated Persistent Spectral Hole-Burning," Proceedings of the International Symposium on Optical Memory 1989, *Japan. J. Appl. Phys.* **28** (Suppl. 28-3), 221 (1989).
34. W. E. Moerner, L. Kador, and W. P. Ambrose, "Ultrasensitive Laser Spectroscopy in Solids: Optical Detection of a Single Dopant Molecule," IEEE Lasers and Electro-Optics Society Annual Meeting Proceedings, IEEE Cat. No. 89CH2641-9, p. 260 (1989).
35. W. E. Moerner, L. Kador, and W. P. Ambrose, "Ultrasensitive Laser Spectroscopy in Solids: Single-Molecule Detection," Proceedings of the Fourth International Conference on Unconventional Photoactive Solids, The Almaden Symposium, *Molec. Cryst. Liq. Cryst.* **183**, 47 (1990).
36. W. E. Moerner, "Persistent Spectral Hole-Burning: Photon-Gating and Fundamental Statistical Limits," in Proceedings of the International Symposium on Polymers for Microelectronics, Science, and Technology (PME '89), Y. Tabata, I. Mita, and S. Nonogaki, editors (Kodansha Scientific and VCH Publishers, 1990), pp. 465-479.
37. W. P. Ambrose and W. E. Moerner, "Temperature Dependence of Photon-Gated Persistent Spectral Hole-Burning for the meso-tetra-p-tolyl-Zn-tetrabenzoporphyrin/chloroform System in poly(Methylmethacrylate)," *Chem. Phys.* **144**, 71 (1990).
38. S. Ducharme, W. P. Risk, W. E. Moerner, V. Y. Lee, R. J. Twieg, and G. C. Bjorklund, "Intracavity Frequency Doubling of a Nd: YAG Laser with an Organic Nonlinear Optical Crystal," *Appl. Phys. Lett.* **57**, 537 (1990).
39. W. E. Moerner, "Ultrasensitive Laser Spectroscopy in Solids: Statistical Fine Structure and Single-Molecule Detection," *New J. Chem.* **15**, 199 (1991).
40. W. P. Ambrose and W. E. Moerner, "Phase-Sensitive Optical Detection of Ballistic Phonon Heat Pulses Using Frequency-Modulation Spectroscopy and Persistent Spectral Holes," *Phys. Rev. B* **43**, 1743 (1990).
41. W. P. Ambrose and W. E. Moerner, "Observation of Spectral Diffusion in Crystals Using Single Impurity Molecules," *Nature* **349**, 225 (1991).
42. W. E. Moerner and W. P. Ambrose, "Comment on 'Single Pentacene Molecules Detected by Fluorescence Excitation in a p-Terphenyl Crystal,'" *Phys. Rev. Lett.* **66**, 1376 (1991).
43. S. Ducharme, J. C. Scott, R. J. Twieg, and W. E. Moerner, "Observation of the Photorefractive Effect in a Polymer," *Phys. Rev. Lett.* **66**, 1846 (1991).

44. W. E. Moerner and W. P. Ambrose, "Finding a Single Molecule in a Haystack: Laser Spectroscopy of Solids from  $\sqrt{N}$  to  $N = 1$ ," *Proc. SPIE* **1435**, 244 (1991).
45. W. P. Ambrose, T. Basché, and W. E. Moerner, "Detection and Spectroscopy of Single Pentacene Molecules in a *p*-Terphenyl Crystal by Means of Fluorescence Excitation," *J. Chem. Phys.*, **95**, 7150 (1991).
46. J. C. Scott, S. Ducharme, R. J. Twieg, and W. E. Moerner, "The Photorefractive Effect in Non-Linear Polymers Doped with Charge Transport Agents," *Polymer Preprints* **32**, 107 (1991).
47. S. Ducharme, J. C. Scott, R. J. Twieg, and W. E. Moerner, "Reply to Comment on Observation of the Photorefractive Effect in a Polymer," *Phys. Rev. Lett.* **67**, 2589 (1991).
48. W. E. Moerner, C. Walsh, J. C. Scott, S. Ducharme, D. M. Burland, G. C. Bjorklund, and R. J. Twieg, "Photorefractivity in Doped Nonlinear Organic Polymers," *Proc. SPIE NLO IV* **1560**, 278 (1991).
49. W. P. Ambrose, Th. Basché, and W. E. Moerner, "Single Molecule Spectral Diffusion in a Solid Detected Via Fluorescence Spectroscopy," *J. Lumin.* **53**, 62 (1992).
50. Th. Basché and W. E. Moerner, "Optical Modification of a Single Impurity Molecule in a Solid," *Nature* **335**, 355 (1992).
51. Th. Basché, W. P. Ambrose, and W. E. Moerner, "Optical Spectra and Kinetics of Single Impurity Molecules in a Polymer: Spectral Diffusion and Persistent Spectral Hole-Burning," *J. Opt. Soc. Amer. B* **9**, 829 (1992).
52. C. A. Walsh and W. E. Moerner, "Two-Beam Coupling Measurements of Grating Phase in a Photorefractive Polymer," to appear in *J. Opt. Soc. Amer. B* (1992).
53. Th. Basché, W. E. Moerner, M. Orrit, and H. Talon, "Photon Antibunching in the Fluorescence of a Single Dye Molecule Trapped in a Solid," *Phys. Rev. Lett.* to appear (1992).
54. W. E. Moerner, "Optical Spectroscopy of Single Molecules in Solids," *Angew. Chemie* (in press, 1992).