

COPY

21

AD-A209 946

OFFICE OF NAVAL RESEARCH
Contract N00014-86-K-0043
TECHNICAL REPORT No. 103

Extraordinary Behavior of Atoms Near a Phase Conjugator

by
Henk F. Arnoldus and Thomas F. George

Prepared for Publication
in
Coherence and Quantum Optics VI
Edited by L. Mandel and E. Wolf
Plenum Press, New York, 1989

Departments of Chemistry and Physics
State University of New York at Buffalo
Buffalo, New York 14260

June 1989

DTIC
ELECTE
JUN 28 1989
S D S D

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 unlimited.

89 6 27 051

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
1a. REPORT SECURITY CLASSIFICATION Unclassified			1b. RESTRICTIVE MARKINGS		
2a. SECURITY CLASSIFICATION AUTHORITY			3. DISTRIBUTION / AVAILABILITY OF REPORT Approved for public release; distribution unlimited		
2b. DECLASSIFICATION / DOWNGRADING SCHEDULE					
4. PERFORMING ORGANIZATION REPORT NUMBER(S) UBUFFALO/DC/TR-103			5. MONITORING ORGANIZATION REPORT NUMBER(S)		
6a. NAME OF PERFORMING ORGANIZATION Depts. Chemistry & Physics State University of New York		6b. OFFICE SYMBOL (if applicable)	7a. NAME OF MONITORING ORGANIZATION		
6c. ADDRESS (City, State, and ZIP Code) Fronczak Hall, Amherst Campus Buffalo, New York 14260			7b. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217		
8a. NAME OF FUNDING / SPONSORING ORGANIZATION Office of Naval Research		8b. OFFICE SYMBOL (if applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER Contract N00014-86-K-0043		
8c. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217			10. SOURCE OF FUNDING NUMBERS		
			PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.
			WORK UNIT ACCESSION NO.		
11. TITLE (Include Security Classification) Extraordinary Behavior of Atoms Near a Phase Conjugator					
12. PERSONAL AUTHOR(S) Henk F. Arnoldus and Thomas F. George					
13a. TYPE OF REPORT		13b. TIME COVERED FROM _____ TO _____		14. DATE OF REPORT (Year, Month, Day) June 1989	
15. PAGE COUNT 4					
16. SUPPLEMENTARY NOTATION Prepared for publication in <u>Coherence and Quantum Optics VI</u> , Edited by L. Mandel and E. Wolf, Plenum Press, New York, 1989					
17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)		
FIELD	GROUP	SUB-GROUP	PHASE CONJUGATOR; ATOM LIFETIME		
			THEORETICAL STUDY TRANSITION DIPOLE GROUND-STATE FLUORESCENCE, (10)		
19. ABSTRACT (Continue on reverse if necessary and identify by block number)					
<p>An atom near a phase conjugator (PC) is theoretically seen to behave quite differently than an atom in empty space or an atom near an ordinary (linear) surface. With two laser beams which pump the nonlinear crystal (PC), it is found that an atom in its ground state can fluoresce if it is sufficiently close to a PC. The possible mechanism involves the atomic transition dipole polarizing the PC, which then spontaneously emits a photon focused on the atom. With stimulated absorption and subsequent spontaneous emission, the net effect is that the atom remains in its ground state. <i>Keywords:</i></p>					
20. DISTRIBUTION / AVAILABILITY OF ABSTRACT <input checked="" type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT. <input type="checkbox"/> DTIC USERS			21. ABSTRACT SECURITY CLASSIFICATION Unclassified		
22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. David L. Nelson			22b. TELEPHONE (Include Area Code) (202) 696-4410		22c. OFFICE SYMBOL

EXTRAORDINARY BEHAVIOR OF ATOMS NEAR A PHASE CONJUGATOR

Henk F. Arnoldus
 Department of Physics
 Villanova University
 Villanova, Pennsylvania 19085

Thomas F. George
 Departments of Chemistry and Physics
 State University of New York at Buffalo
 Buffalo, New York 14260

An atom near a phase conjugator (PC) behaves quite differently than an atom in empty space, or an atom in the vicinity of an ordinary (linear) surface. A spherical diverging wave, which is emitted by the atomic dipole $\underline{\mu}$, is reflected by the PC as a converging wave, and exactly focused on the emitting atom. This gives rise to a high probability for reabsorption of the photon that was just emitted. It can be anticipated that this mechanism leads to a strong enhancement of the effective lifetime of an excited state. We have calculated the spatial intensity distribution of the fluorescence, which is emitted in the half space to the 'left' of the PC (Fig. 1), and the total emitted power

$$P(t) = 2\epsilon_0 c r^2 \int d\Omega \langle \underline{E}^{(-)}(\underline{r}, t) \cdot \underline{E}^{(+)}(\underline{r}, t) \rangle \quad (1)$$

in the radiation zone. The angle brackets denote a quantum average, and $\underline{E}^{(+)}(\underline{r}, t)$ is the positive-frequency part of the electric field operator in the Heisenberg picture. This $\underline{E}^{(+)}$ is the solution of Maxwell's equations for this configuration, and it includes both the emitted field by the dipole and the field which is reflected by the PC. It is assumed that the PC is of the four-wave mixing type.

For a degenerate two-level atom we find the general expression

$$P(t) = \frac{\omega_0^4}{6\pi\epsilon_0 c^3} \text{Tr} \rho(t-r/c) \{ \underline{\mu}^{(-)} \cdot \underline{\mu}^{(+)} + c_{\parallel} \underline{\mu}_{\parallel}^{(+)} \cdot \underline{\mu}_{\parallel}^{(-)} + c_{\perp} \underline{\mu}_{\perp}^{(+)} \cdot \underline{\mu}_{\perp}^{(-)} \} \quad (2)$$

where (+) indicates the lowering part of the dipole operator (Schrödinger picture). Here, $\rho(t)$ is the density operator of the atom only, and ω_0 is the atomic transition frequency. The two c-coefficients are found to be [1]

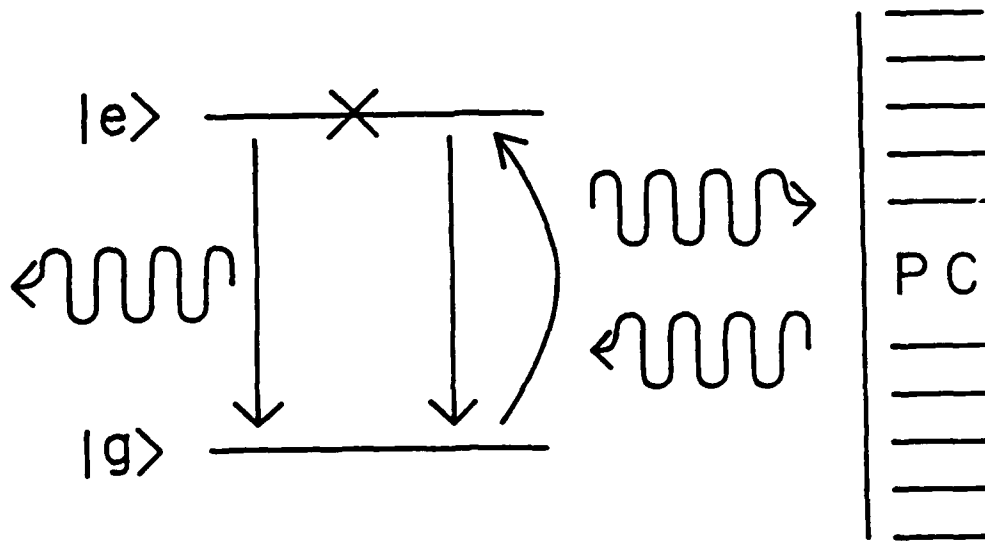


Fig. 1. Illustration of the possible mechanisms which give rise to the first term on the right-hand side of Eq. (4). A photon emitted to the left can be detected, but a photon that hits the PC never reaches the detector. This is either because this photon is absorbed by the medium, or it is absorbed by the atom after reflection. The figure depicts the second mechanism.

$$c_{\parallel} = \frac{3}{4} \int_0^1 du (1+u^2) |R(\omega_0, u)|^2, \quad c_{\perp} = \frac{3}{2} \int_0^1 du (1-u^2) |R(\omega_0, u)|^2, \quad (3)$$

where R is the Fresnel coefficient for reflection of a plane wave with the cosine of the angle of incidence on the surface of the PC equal to u . These coefficients also depend on the atomic level separation ω_0 , in general. In the case that the dependence of the Fresnel coefficients on the angle of incidence can be neglected, both c -coefficients reduce to $|R|^2$.

Expression (2) can be simplified considerably if we assume that the atom has only two states, and that R varies only slightly with the angle of incidence. Then Eq. (2) reduces to

$$P(t) = \frac{1}{2} A \hbar \omega_0 [n_e(t-r/c) + |R|^2 n_g(t-r/c)], \quad (4)$$

with A as the Einstein coefficient for spontaneous decay in empty space, and n_e and n_g as the populations of the excited and ground state, respectively. Nothing has been assumed about the time dependence of these populations, and so Eq. (4) holds also when the atom is not in its steady state.

Result (4) has two remarkable features. First, we notice that the first term, which is proportional to n_e , is just one half times the emission rate of an atom in empty space. This implies that the photons which are emitted towards the PC, and subsequently reflected (Fig. 1), are reabsorbed completely. No reflected photon passes the atom. We would like to emphasize, however, that this is only one of the tenable interpretations of the first term in Eq. (4). It is also possible that the PC absorbs the photon which is incident on its surface completely, and without reflection. This process would leave the atom in its ground state, rather than in its excited state, after occurrence of this process. Another possibility would be that both processes can happen.

Secondly, we see that $P(t)$ has a term proportional to the population of the ground state. From this we conclude that an atom in its ground state can fluoresce, if it is sufficiently close to a PC. The possible mechanism is illustrated in Fig. 2. The presence of the atomic dipole moment polarizes the PC, which then spontaneously emits a photon, focused on the atom. Stimulated absorption and subsequent spontaneous emission gives rise to an observable photon at the left. The net effect is that the atom remains in its ground state. The required energy for this process is of course provided by the two laser beams which pump the nonlinear crystal.

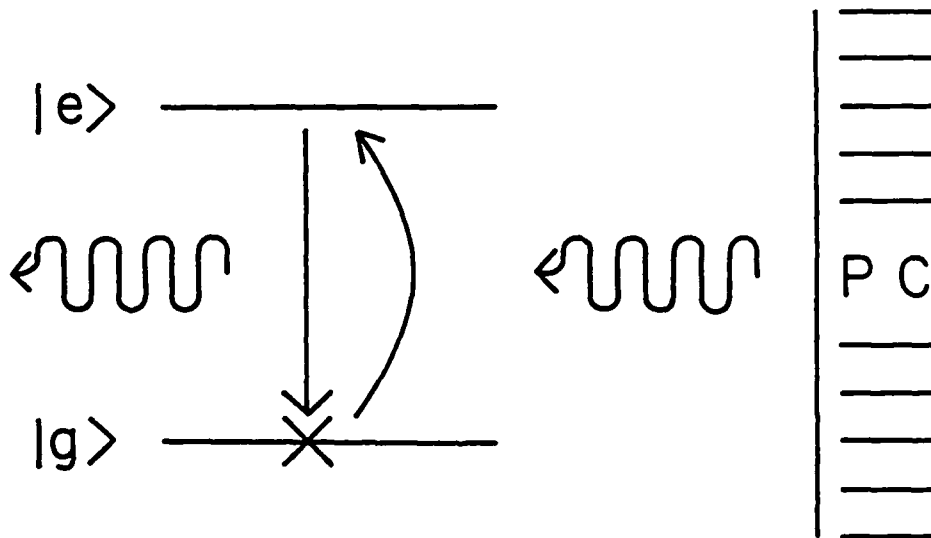


Fig. 2. As predicted by Eq. (4), an atom in its ground state can effectively emit a photon. Due to energy conservation this photon must first be extracted from the PC. The polarized PC spontaneously emits a focused photon, which is absorbed by the atom. Spontaneous excitation and subsequent decay produce a detectable photon at the left. After the process, the atom is again in its ground state.

This work was partially supported by ONR and AFOSR.

[1] H. F. Arnoldus and T. F. George, Opt. Lett. (1989), submitted.



J
□
□
Code

A-1

TECHNICAL REPORT DISTRIBUTION LIST, GEN

	<u>No. Copies</u>		<u>No. Copies</u>
Office of Naval Research Attn: Code 1113 800 N. Quincy Street Arlington, Virginia 22217-5000	2	Dr. David Young Code 334 NORDA NSTL, Mississippi 39529	1
Dr. Bernard Douda Naval Weapons Support Center Code 50C Crane, Indiana 47522-5050	1	Naval Weapons Center Attn: Dr. Ron Atkins Chemistry Division China Lake, California 93555	1
Naval Civil Engineering Laboratory Attn: Dr. R. W. Drisko, Code L52 Port Hueneme, California 93401	1	Scientific Advisor Commandant of the Marine Corps Code RD-1 Washington, D.C. 20380	1
Defense Technical Information Center Building 5, Cameron Station Alexandria, Virginia 22314	12 high quality	U.S. Army Research Office Attn: CRD-AA-IP P.O. Box 12211 Research Triangle Park, NC 27709	1
DTNSRDC Attn: Dr. H. Singerman Applied Chemistry Division Annapolis, Maryland 21401	1	Mr. John Boyle Materials Branch Naval Ship Engineering Center Philadelphia, Pennsylvania 19112	1
Dr. William Tolles Superintendent Chemistry Division, Code 6100 Naval Research Laboratory Washington, D.C. 20375-5000	1	Naval Ocean Systems Center Attn: Dr. S. Yamamoto Marine Sciences Division San Diego, California 91232	1
		Dr. David L. Nelson Chemistry Division Office of Naval Research 800 North Quincy Street Arlington, Virginia 22217	1

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. J. E. Jensen
Hughes Research Laboratory
3011 Malibu Canyon Road
Malibu, California 90265

Dr. J. H. Weaver
Department of Chemical Engineering
and Materials Science
University of Minnesota
Minneapolis, Minnesota 55455

Dr. A. Reisman
Microelectronics Center of North Carolina
Research Triangle Park, North Carolina
27709

Dr. M. Grunze
Laboratory for Surface Science and
Technology
University of Maine
Orono, Maine 04469

Dr. J. Butler
Naval Research Laboratory
Code 6115
Washington D.C. 20375-5000

Dr. L. Interante
Chemistry Department
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. Irvin Heard
Chemistry and Physics Department
Lincoln University
Lincoln University, Pennsylvania 19352

Dr. K.J. Klaubunde
Department of Chemistry
Kansas State University
Manhattan, Kansas 66506

Dr. C. B. Harris
Department of Chemistry
University of California
Berkeley, California 94720

Dr. F. Kutzler
Department of Chemistry
Box 5055
Tennessee Technological University
Cookeville, Tennessee 38501

Dr. D. DiLella
Chemistry Department
George Washington University
Washington D.C. 20052

Dr. R. Reeves
Chemistry Department
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. Steven M. George
Stanford University
Department of Chemistry
Stanford, CA 94305

Dr. Mark Johnson
Yale University
Department of Chemistry
New Haven, CT 06511-8118

Dr. W. Knauer
Hughes Research Laboratory
3011 Malibu Canyon Road
Malibu, California 90265

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. G. A. Somorjai
Department of Chemistry
University of California
Berkeley, California 94720

Dr. J. Murday
Naval Research Laboratory
Code 6170
Washington, D.C. 20375-5000

Dr. J. B. Hudson
Materials Division
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. Theodore E. Madey
Surface Chemistry Section
Department of Commerce
National Bureau of Standards
Washington, D.C. 20234

Dr. J. E. Demuth
IBM Corporation
Thomas J. Watson Research Center
P.O. Box 218
Yorktown Heights, New York 10598

Dr. M. G. Lagally
Department of Metallurgical
and Mining Engineering
University of Wisconsin
Madison, Wisconsin 53706

Dr. R. P. Van Duyne
Chemistry Department
Northwestern University
Evanston, Illinois 60637

Dr. J. M. White
Department of Chemistry
University of Texas
Austin, Texas 78712

Dr. D. E. Harrison
Department of Physics
Naval Postgraduate School
Monterey, California 93940

Dr. R. L. Park
Director, Center of Materials
Research
University of Maryland
College Park, Maryland 20742

Dr. W. T. Peria
Electrical Engineering Department
University of Minnesota
Minneapolis, Minnesota 55455

Dr. Keith H. Johnson
Department of Metallurgy and
Materials Science
Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

Dr. S. Sibener
Department of Chemistry
James Franck Institute
5640 Ellis Avenue
Chicago, Illinois 60637

Dr. ^{Arnold} Arnold Green
Quantum Surface Dynamics Branch
Code 3817
Naval Weapons Center
China Lake, California 93555

Dr. A. Wold
Department of Chemistry
Brown University
Providence, Rhode Island 02912

Dr. S. L. Bernasek
Department of Chemistry
Princeton University
Princeton, New Jersey 08544

Dr. W. Kohn
Department of Physics
University of California, San Diego
La Jolla, California 92037

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. F. Carter
Code 6170
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. Richard Colton
Code 6170
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. Dan Pierce
National Bureau of Standards
Optical Physics Division
Washington, D.C. 20234

Dr. R. Stanley Williams
Department of Chemistry
University of California
Los Angeles, California 90024

Dr. R. P. Messmer
Materials Characterization Lab.
General Electric Company
Schenectady, New York 22217

Dr. Robert Gomer
Department of Chemistry
James Franck Institute
5640 Ellis Avenue
Chicago, Illinois 60637

Dr. Ronald Lee
R301
Naval Surface Weapons Center
White Oak
Silver Spring, Maryland 20910

Dr. Paul Schoen
Code 6190
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. John T. Yates
Department of Chemistry
University of Pittsburgh
Pittsburgh, Pennsylvania 15260

Dr. Richard Greene
Code 5230
Naval Research Laboratory
Washington, D.C. 20375-5000

Dr. L. Kesmodel
Department of Physics
Indiana University
Bloomington, Indiana 47403

Dr. K. C. Janda
University of Pittsburgh
Chemistry Building
Pittsburg, PA 15260

Dr. E. A. Irene
Department of Chemistry
University of North Carolina
Chapel Hill, North Carolina 27514

Dr. Adam Heller
Bell Laboratories
Murray Hill, New Jersey 07974

Dr. Martin Fleischmann
Department of Chemistry
University of Southampton
Southampton SO9 5NH
UNITED KINGDOM

Dr. H. Tachikawa
Chemistry Department
Jackson State University
Jackson, Mississippi 39217

Dr. John W. Wilkins
Cornell University
Laboratory of Atomic and
Solid State Physics
Ithaca, New York 14853

ABSTRACTS DISTRIBUTION LIST, 056/625/629

Dr. R. G. Wallis
Department of Physics
University of California
Irvine, California 92664

Dr. D. Ramaker
Chemistry Department
George Washington University
Washington, D.C. 20052

Dr. J. C. Hemminger
Chemistry Department
University of California
Irvine, California 92717

Dr. T. F. George
Chemistry Department
University of Rochester
Rochester, New York 14627

Dr. G. Rubloff
IBM
Thomas J. Watson Research Center
P.O. Box 218
Yorktown Heights, New York 10598

Dr. Horia Metiu
Chemistry Department
University of California
Santa Barbara, California 93106

Dr. W. Goddard
Department of Chemistry and Chemical
Engineering
California Institute of Technology
Pasadena, California 91125

Dr. P. Hansma
Department of Physics
University of California
Santa Barbara, California 93106

Dr. J. Baldeschwieler
Department of Chemistry and
Chemical Engineering
California Institute of Technology
Pasadena, California 91125

Dr. J. T. Keiser
Department of Chemistry
University of Richmond
Richmond, Virginia 23173

Dr. R. W. Plummer
Department of Physics
University of Pennsylvania
Philadelphia, Pennsylvania 19104

Dr. E. Yeager
Department of Chemistry
Case Western Reserve University
Cleveland, Ohio 44106

Dr. N. Winograd
Department of Chemistry
Pennsylvania State University
University Park, Pennsylvania 16802

Dr. Roald Hoffmann
Department of Chemistry
Cornell University
Ithaca, New York 14853

Dr. A. Stecki
Department of Electrical and
Systems Engineering
Rensselaer Polytechnic Institute
Troy, New York 12181

Dr. G.H. Morrison
Department of Chemistry
Cornell University
Ithaca, New York 14853