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The search for new optical materials for spectral hole burning applications, evaluation of their dynamic and static properties, and studies of the ultimate limits on material performance consistent the research that was carried out. We strive to understand the atomic-scale mechanisms that determine material performance. Attention is focused on rare earth and transition metal ion materials; systems for photon-gated hole burning are explored. General facilities for hole burning research are available in our laboratory, and we have fifteen years experience in this area, studying a variety of rare earth activated materials.

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**FINAL
Annual Report**

for year September 1, 1995 to ^{December}~~August~~ 31, 1996

submitted by

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on

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**SPECTROSCOPY OF MATERIALS FOR PERSISTENT SPECTRAL
HOLE BURNING OPTICAL MEMORIES AND SIGNAL PROCESSING**

submitted to

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Objectives

The search for new optical materials for spectral hole burning applications, evaluation of their dynamic and static properties, and studies of the ultimate limits on material performance constitute the research that we carry out under the *AFOSR Physics and Electronics Sciences Program of Dr. Alan E. Craig*. We strive to understand the atomic-scale mechanisms that determine material performance. Attention is focused on rare earth and transition metal ion materials; systems for photon-gated hole burning are explored. General facilities for hole burning research are available in our laboratory, and we have fifteen years experience in this area, studying a variety of rare earth activated materials.

Our work at Montana State University is carried out in collaboration with *Scientific Materials Corporation -- an AFOSR SBIR Phase II contractor*, with Roger Macfarlane at IBM Almaden Research Center by a subaward under this grant, with groups at the University of Oxford, UK, and with other groups in the US and France. Additional support for this work at Montana State University has been provided by NASA-EPSCoR, NSF-EPSCoR, and NATO Collaborative Research grants.

Status of Effort

Our research on persistent spectral hole burning and optical coherent transients is directed toward

- a) studies of atomic-scale material properties and interaction mechanisms that establish limits on material performance in applications including optical storage and high speed signal processing -- work focused on new Tm^{3+} doped materials,
- b) design, evaluation, and development of new materials -- a number of new Tm^{3+} compounds were developed including $\text{Tm}^{3+}:\text{YGG}$ and disordered $\text{Tm}^{3+}:\text{YLuAG}$,
- c) "photon gated" spectral hole burning -- work focused on search for new ions such as Tb^{3+} and new hosts,
- d) new materials for use with diode lasers -- both Tm^{3+} materials for 790 nm and materials for the 1.5 μm communications region,
- e) provision of solid state frequency references for stable lasers and optical clocks in communications and computer systems using persistent spectral hole burning.
- f) development of materials for optical routers based on spatial-spectral holography.

We participated in two AFOSR Workshops,

- a) "Hole Burning Materials and Physical Processes, R. L. Cone," Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, March 3-6, 1996.
- b) "Spectral-Hole-Burning Frequency Standards and Stable Lasers," R. L. Cone, Contractors Workshop on Time and Frequency Standards, Phillips Laboratory, Kirtland Air Force Base, Albuquerque, NM, April 25, 1996.

Accomplishments/New Findings

Particular effort during this grant period was concentrated on Tm^{3+} compounds for use with semiconductor diode lasers at 790 nm.

Studies of $\text{Tm}^{3+}:\text{Y}_2\text{SiO}_5$ and $\text{Tm}^{3+}:\text{Y}_2\text{Si}_2\text{O}_7$ for memory, signal-processor and router applications were completed and a manuscript published in Optics Letters.

A number of new Tm^{3+} materials were designed and evaluated for these applications. Included were $\text{Tm}:\text{YGG}$ (yttrium gallium garnet), $\text{Tm}:\text{YLuAg}$ (yttrium-lutecium aluminum garnet), $\text{Tm}:\text{Y}_2\text{O}_3$ (yttria), and $\text{Tm}:\text{LuAg}$ (lutecium aluminum garnet).

- a) For $\text{Tm}:\text{YGG}$, a significant increase in homogeneous linewidth to 60 GHz was achieved while maintaining a very favorable ratio of inhomogeneous to homogeneous linewidths of $\Gamma_{\text{inh}}/\Gamma_{\text{h}} = 8 \times 10^7$; at the same time the effects of instantaneous diffusion (laser-induced dephasing) were eliminated for "one color" experiments; that dramatic result is being investigated further with a new batch of crystals.
- b) The related compound $\text{Tm}:\text{YLuAg}$ was designed for increased signal bandwidth capability by intentionally introducing disorder in the occupation of the yttrium sites with a 50:50 mix of Y^{3+} and Lu^{3+} ; this produced the substantially increased inhomogeneous broadening as expected, with no penalty on the ratio of inhomogeneous to homogeneous linewidths of $\Gamma_{\text{inh}}/\Gamma_{\text{h}} = 6 \times 10^7$. This extends the potential signal processing bandwidth at semiconductor laser wavelengths to 270 GHz. Remarkably, this material functioned well at 3% Tm concentration, giving strong absorption that will allow thin media (2 mm) for storage or processing.
- c) For $\text{Tm}:\text{Y}_2\text{O}_3$, there was evidence of structural disorder on the oxygen sites. Our photon echo measurements can be used to very sensitively assess yttria materials.
- d) In $\text{Tm}:\text{LuAg}$, we again found that operation at high Tm concentration was possible. Again, this warrants further attention with the goal of new insights into instantaneous spectral diffusion (laser-induced dephasing).

With these materials we have again established new ranges of parameter space for device applications with semiconductor lasers -- they could handle signal bandwidths as high as 270 GHz.

We are exploring new materials and materials concepts for photon gating of persistent spectral hole burning. The "robustness" and long-term persistence associated with photon gating are important to storage and processing and to frequency standard and optical clock applications.

- a) In conjunction with Scientific Materials, we are designing and evaluating co-doped materials with paired sites capable of stabilizing ions in multiple ionization states. Co-doping of active ions that may be stabilized in more than one ionization state may overcome efficiency problems for gating and may also reduce the effect of gating on the surrounding area in the crystal and hence reduce destruction of previously written data by spectral diffusion.
- b) To extend knowledge of photon gating, we have been working on materials where photoionization of Tb^{3+} to Tb^{4+} can open new areas of study.

Among the general material device parameters being studied are

- a) limits on the optical homogeneous line widths (population lifetime T_1 , crystal field splittings, phonons, crystal lattice composition [structure and disorder], and laser power-induced instantaneous spectral diffusion),
- b) spin-lattice relaxation and spectral diffusion processes affecting spectral-hole lifetime,
- c) limits on hole-writing and reading rates,
- d) lifetimes of metastable intermediate states important for population storage, for example in Tm^{3+} compounds,
- e) concentration-dependent dephasing (instantaneous spectral diffusion),
- f) growth-dependent effects arising from oxygen vacancies and paramagnetic impurities,
- g) material band gaps and position of ionic ground and excited states within the band gap for designing photon gating strategies (by ultraviolet and x-ray photoemission spectroscopy), and
- h) two-laser photon gating efficiencies and hole widths.

We continue to study samples from Scientific Materials Corporation of Bozeman, IBM (Roger Macfarlane), several groups at AT&T Bell Laboratories, Yale (W.P. Wolf, S. Mroczkowski), U. Lyon-France (B. Jacquier), Lawrence Livermore National Laboratory (M.J. Weber), Lawrence Berkeley Laboratory (N.M. Edelstein), M.I.T., CREOL, and others.

Several unusual materials doped with rare earth and transition metal ions have been obtained.

Personnel Supported and Associated

Dr. Yongchen Sun arrived July 1, 1996, from the University of Georgia, for a two-year postdoctoral appointment as "Assistant Research Professor."

Dr. Flurin Koenz won a postdoctoral fellowship from the Swiss National Science Foundation specifically for research in Cone's laboratory. Dr. Koenz arrives from Bern, Switzerland, on October 14, 1996, with all expenses paid by the Swiss NSF.

A third postdoctoral fellow, Nicholas Strickland from the group of Dr. Glynn Jones at University of Canterbury, Christchurch, New Zealand, will join Cone's group in Autumn, 1996. The Glynn Jones' group is well known for its work on persistent spectral hole burning in H- and D-doped CaF_2 rare earth materials and for general expertise in rare earth spectroscopy and modeling.

Cone's group has had four graduate students involved in AFOSR research this past year. Their salaries were paid by other research grants.

- 1) Guangming Wang, graduate student in physics, supported by NASA-EPSCoR.
- 2) Greg White, graduate student in physics, supported by NSF-EPSCoR and Montana Space Grant Fellowship.
- 3) Lane Seeley, graduate student in physics, supported by NSF-EPSCoR.
- 4) Todd Harris, graduate student in physics, supported by NSF-EPSCoR.

Cone's group has had two undergraduate students involved in AFOSR research this past year.

- 1) Charles Thiel, undergraduate student in physics, BS in Physics, May, 1996, and a Congressional Goldwater Scholar. Charles will continue for a Ph.D. at Montana State University.
- 2) Tyler Morgus, undergraduate student in physics, BS in Physics, May, 1996. Tyler will enter the Ph.D. program at Lehigh University in September, 1996.

Dr. M.J.M. Leask of the Clarendon Laboratory, University of Oxford, visited Cone's group three times during this grant year:

- a) October 4 - December 4, 1995, while on sabbatical leave,
- b) February 29 - March 7, 1996, for discussions, manuscript writing, and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning,
- c) August 6 - 20, 1996, to continue this work and for experiments on rare-earth nanocrystals in carbon nanotubes prepared at the University of Oxford.

Dr. Roger Macfarlane of IBM Almaden Research Center visited March 3 - March 11, 1996, for discussions, experiments, and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning.

There was regular communication with both the IBM and Oxford groups throughout the year.

Dr. Yongchen Sun of the University of Georgia Department of Physics visited February 29 - March 7, 1996, for discussions and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning, in preparation to assume a postdoctoral appointment here as "Assistant Research Professor."

Dr. Richard Meltzer of the University of Georgia visited February 29 - March 7, 1996, for discussions and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning.

Dr. William Randall Babbitt of the University of Washington visited in May, 1995, and presented a Physics Colloquium. Babbitt visited again on March 6-8 to discuss a joint University of Washington - Montana State University proposal on a prototype terabit/second optical fiber communications link using spectral hole burning concepts and frequency-division multiplexing. This proposal was submitted to the ARPA-Ultra-Photonics program in June, 1996.

Hai Lin of the University of Oregon, Jean-Pierre Galaup of Paris, Aleksander Rebane of ETH-Zurich, and Yiping Zhang of SRI, Menlo Park, toured Rufus Cone's research laboratory in conjunction with the Big Sky Workshop on Applications of Persistent Spectral Hole Burning.

Dr. Bernard Jacquier from Universite Lyon-I visited July 27-August 3, 1996, to discuss hole burning in rare earth doped optical waveguides and dielectric microcavities.

The interdisciplinary MSU Optical Technology Center (OpTeC) group submitted an NSF Optical Science and Engineering proposal on hole burning memory materials. This group consists of Cone and Carlsten from Physics, Singel and Spangler from Chemistry, and Cady from Electrical Engineering. Dr. Aleksander Rebane has accepted a position in the group.

Publications

1. "Optical Dephasing Mechanisms in $\text{Tm}^{3+}:\text{Y}_2\text{Si}_2\text{O}_7$," G. M. Wang, R. W. Equall, R. L. Cone, M. J. M. Leask, K. W. Godfrey, and F. R. Wondre, *Opt. Lett.* **21**, 818 (1996).
2. "Homogeneous Broadening of Optical Transitions in $\text{Tm}^{3+}:\text{Y}_2\text{SiO}_5$," R. W. Equall, G. M. Wang, R. L. Cone, and R. M. Macfarlane, to be submitted to *Phys. Rev. B*.
3. "Spectral Hole Burning and ODNQR in Flux-Grown Stoichiometric Europium Vanadate Crystals," P. C. Hansen, M. J. M. Leask, B. M. Wanklyn, Y. Sun, R. L. Cone, and M. M. Abraham, submitted to *Phys. Rev. B.*, August 19, 1996.

Interactions

Presentations and Participation at Meetings and Conferences

"New Materials for Persistent Spectral Hole Burning Optical Memories and Signal Processing Applications," R. W. Equall, MSU Physics Department Colloquium, October 20, 1995.

R. L. Cone attended the Advanced Solid-State Lasers meeting, San Francisco, CA, January 31-February 2, 1996.

R. L. Cone attended the American Physical Society 1996 March Meeting, St. Louis, MO, March 18-22, 1996.

Invited workshop talk, "Hole Burning Materials and Physical Processes, R. L. Cone," AFOSR Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, March 3-6, 1996.

Invited workshop talk, "Spectral-Hole-Burning Frequency Standards and Stable Lasers," R. L. Cone, AFOSR Contractors Workshop on Time and Frequency Standards, Phillips Laboratory, Kirtland Air Force Base, Albuquerque, NM, April 25, 1996.

Program Committee, Conference on Lasers and Electro-Optics (CLEO/QUELS '96), Anaheim, CA, June 2-7, 1996.

Presider, Session on Inorganic Optical Materials, Conference on Lasers and Electro-Optics (CLEO/QUELS '96), Anaheim, CA, June 2-7, 1996.

"Tm Materials for Coherent Transient Optical Memories and Signal Processors," R. W. Equall, G. M. Wang, G. A. White, R. L. Cone, M. J. M. Leask, R. M. Macfarlane, and R. L. Hutcheson, Conference on Lasers and Electro-Optics (CLEO/QUELS '96), Anaheim, CA, June 2-7, 1996.

Invited conference talk, "Nonlinear Optical Studies of Rare Earth Compounds for Optical Storage and Signal Processing and for Lasers," R. L. Cone, 21st Rare Earth Research Conference, Duluth, MN, July 7-12, 1996.

Invited conference talk, "Photon Echo Measurements of Optical Dephasing in Rare-Earth Doped Insulators," R. L. Cone, International Conference on Luminescence, Prague, Czech Republic, August 18-23, 1996.

Talks at Montana State University "OpTeC" Conference on Optical Science and Laser Technology, Bozeman, MT, Sept. 5-6, 1995.

"Rare Earth Crystals for Solid State Lasers and for Optical Data Storage and Signal Processing," R. L. Cone.

"Tm³⁺-Doped Materials for Optical Memory and Signal Processing Applications," Guangming Wang, graduate student.

"Materials for Photon Gated Hole Burning," G. White, graduate student.

"Searches for Tb Compounds Exhibiting Photon Gated Hole Burning," C. W. Thiel, undergraduate student.

Consultative and Advisory Functions

Cone's group served as advisors on crystal design and characterization to Scientific Materials Corporation, an AFOSR SBIR Phase II contractor. They also worked to enhance linkage of Scientific Materials to other groups in the Spectral Hole Burning community.

A proposal on optical routers for fiber optical systems with terabit bandwidths, more than two orders of magnitude beyond the current state of the art, was prepared and submitted by W. Randall Babbitt of the University of Washington and the Cone group to the ARPA Ultra-Photonics program in June, 1996.

Dr. Ralph Kelley of the Atomic Physics Section of AFOSR invited Cone to participate in a Workshop on Time and Frequency Standards on April 25. A talk entitled "Spectral Hole Burning Frequency Standards and Stable Lasers" will be presented at this meeting based on our narrow line spectroscopy, our goals for gated materials, and John's work on laser frequency locking. The other participants were Selim Shahriar and Philip Hemmer of Hanscom Air Force Base; Ronald Walsworth, Smithsonian Astrophysical Observatory - Harvard; Daniel Kleppner, Physics Department, MIT; Gerald Gabrielse, Physics Department Lyman Laboratory - Harvard; Steve Chu, Physics Department, Stanford; and J.L. Hall, JILA-Boulder.

Transitions

Materials developed and characterized by Cone's group are being used by a number of other groups in the Spectral Hole Burning Applications community to develop demonstration optical memory and signal processing devices.

Crystals of Tm^{3+} :YGG (yttrium gallium garnet) grown by Scientific Materials, with our advice and characterization, are currently being used by several groups to significantly improve device performance over Tm^{3+} :YAG.

Cone is negotiating a material development program with the Los Alamos Group that is working on laser-induced cooling, a technique that may provide the means for practical operation of optical memory or processing devices based on PSHB at lower temperatures.

Development and evaluation of new materials is a special role that we assume in the *AFOSR Photonic Devices and Systems Program of Dr. Alan E. Craig*. With Scientific Materials we are actively participating in the design of the materials. We cooperatively optimize the material properties through the close interaction with Scientific Materials made possible by our location; by providing rapid feedback, adjustments may be made to the synthesis process, leading to development of better quality materials. Characterized samples are forwarded to other groups funded by AFOSR with the specific mission to use them to develop designs for practical devices.

New Discoveries, Inventions, or Patent Disclosures

The technical progress on materials for use with semiconductor lasers, reported above in "Status of Effort" and "Accomplishments," was made possible by the design and characterization of a number of new Tm^{3+} materials including Tm :YGG (yttrium gallium garnet) and Tm :YLuAg (yttrium-lutecium aluminum garnet).

- a) In Tm :YGG, a significant increase in homogeneous linewidth to 60 GHz was achieved while at the same time reducing the effects of instantaneous diffusion (laser-induced dephasing).
- b) In Tm :YLuAg, intentional introduction of disorder in the occupation of the yttrium lattice sites produced substantial inhomogeneous broadening without loss of narrow homogeneous linewidths; as a result potential signal processing bandwidth at semiconductor laser wavelengths is extended to 270 GHz. Remarkably, this material functioned well at 3% Tm concentration, making possible thin storage or processing media (2 mm).

A patent application is in preparation.

Honors and Awards

Principal Investigator Rufus L. Cone received the Cox Family Award for Creative Scholarship and Teaching from the Montana State University Foundation, May, 1996. In consecutive years Cone has received MSU's top two academic and research awards.

The Graduate Achievement Award at Montana State University was presented to Randy Wayne Equall for his Ph.D. thesis research. The award was presented on May 10, 1996.

During Autumn Semester, 1995, Gregg White was supported by a Montana Space Grant Fellowship.

The Dean's Award for Excellence in the College of Letters and Science was presented to Charles Thiel, an undergraduate working in this group. Charles is a Congressional Barry M. Goldwater Scholar and will enter graduate study at MSU in the Autumn, 1996. Charles continued his gated spectral hole burning research during Summer, 1996.

Tyler Morgus, an undergraduate physics major working in this group, accepted graduate admission to the Ph.D. program at Lehigh University and worked in a research laboratory there during summer, 1996.

Dr. Flurin Koenz of the University of Berne, Switzerland, won a postdoctoral fellowship from the Swiss National Science Foundation for research in Cone's laboratory.