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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS	READ INSTRUCTIONS BEFORE COMPLETING FORM	
REPORT NUMBER	2. GOVT ACCESS	ON NO. 3. RECIPIENT'S CATALOG NUMBER	$1 \cap$	
ARO 21549.147	N/A	N/A	10	
Applications of Nonlinear Hole Burning Spectroscopy		5. TYPE OF REPORT & PERIOD COVERED Final report for period of		
		July 1, 1985 - June 30, 1987		
		5. PERFORMING ORG. REPORT NUMBER		
AUTHOR(a)		. CONTRACT OR GRANT NUMBER(s)	1	
John C. Wright		DAAG29-85-K-0001]	
]	
PERFORMING ORGANIZATION NAME AND ADDRESS		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS		
Department of Chemistry, University of Wisconsin 1101 University Avenue, Madison, Wisconsin 53706				
CONTROLLING OFFICE NAME AND ADDRESS		12. REPORT DATE		
U. S. Army Research Office		August 31, 1987		
rost UIIIce Box 12211 Research Triangle Park, NC 27700		3		
MONITORING AGENCY NAME & ADDRESS(If different from Controlling Office)		ttice) 15. SECURITY CLASS. (of this report)		
		Unclassified		
		154. DECLASSIFICATION/DOWNGRADING SCHEDULE		
OSTRIBUTION STATEMENT (of this Report) Approved for public release;	distribution un]	limited. DTIC ELECTE		
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ARO 21549.1-84

Applications of Nonlinear Hole Burning Spectroscopy

Final Report

John C. Wright

August 31, 1987

U.S. Army Research Office

DAAG29-85-K-0001

University of Wisconsin

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This research program was directed toward the demonstration of feasibility for a new type of spectral hole burning that would use nonlinear four wave mixing to generate an excited state population. Upon relaxation, the excited population would benerate a spectral hole through the generation of phonons interacting with the local environment of the surrounding glass. The key advantage to this approach would be the tremendous increase in information density achievable with four wave mixing because all of the intermediate resonances would also be accessed in the four wave mixing. Since each intermediate level would have an inhomogeneous envelope that did not correlate with the broadening parameters of other transitions, each intermediate level could be used as a label for the information storage.

The first part of the research project involved a search for a suitable host material and an active chromophore in which to attempt the nonlinear hole burning. A number of dopant molecules were tried although we concentrated on porphine ring materials where hole burning has been observed commonly. These included free base porphine, phthalocyanine, porphine, octaethylporphine, tetraphenylporphine, pentacene, and perylene. The host materials included methyl tetrahydrofuran, n-octane, benzoic acid, polymethyl methacrylate, and o-terphenyl. The experiments involved identifying the vibrational, electronic and vibronic levels that would be accessed in the four wave mixing by using fluorescence line narrowing techniques, measuri.g the hole burning efficiency, and performing four wave mixing experiments in the materials. Although our initial studies were discouraging, the latest studies in fact identified an excellent candidate system - polymethyl methacrylate doped with either pentacene or phthalocyanine.

There were also a number of experimental difficulties that had to be addressed in the research. The major difficulty was to have a reliable way to identify the position of the laser wavelengths during a spectral scan. In all of our experiments, two lasers had to be scanned in synchronization with each other so that intermediate resonances were not lost during the scan. Since the resonances were very sharp, the synchronization had to be excellent. We found that mechanical tolerances were not sufficient to maintain the necessary synchronization and that the wavelength needed to be monitored interferometrically. A Fizeau interferometer was used to determine the lasers' wavelength accurately. With this approach, we found that reliable and high quality four wave mixing scans could be acquired.

We have found that holes could be burned in PMMA with a very high efficiency as evidenced by the rapid loss of signal when lasers were resonant with the transition. In order to measure and characterize the holes, it was necessary to perform spectral scans across them. Here we ran into another problem since the hole burning was so efficient. As the spectral scans were performed, new holes were burned that interfered with the hole already created. The efficiency is so high that one could not read out the hole with a fast enough scan or with a low enough intensity. For this reason, we began the development of a new approach to measuring hole burning.

If all of the lasers are scanned so they are constantly accessing new sites, the hole burning can be distributed across the entire inhomogeneous line with a distribution that depends upon the rate of scanning. Two lasers are scanned slowly in synchronization so that a set of sites with a common vibrational level are accessed. In order to obtain information about the sites, a third laser is also scanned but at a more rapid rate. This laser will preferentially pick out the resonances associated with the sites that are resonant with the first two lasers. Sites with deep holes will appear during the scans because they will have a much larger population depletion than the sites resonant only briefly during the scan. The contrast level between the sites is selectable by the scan rate of the first two lasers. This approach is a very new one for the field of hole burning. It opens the possibility for information storage in a series of holes that are determined by the rate of scanning used to burn the holes that are determined by the rate of scanning used to burn the holes in the first place. The technique is being tested using fluorescence line narrowing techniques and has worked as expected. A publication is anticipated on this new technique in the nottoo-distant future. We are optimistic that the use of the same technique with four wave mixing will demonstrate the presence of hole burning with nonlinear spectroscopy.

Publications - None.

Participating Scientific Personnel - Bradford B. Price Bruce K. Winker Gregory B. Hurst John C. Wright

Bibliography - None.

Appendices - None.

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