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13. ABSTRACT *(Maximum 200 words)*

Results of our earlier research in the realm of quantum optics were extended in order to solve the challenging technical problems of importance. Specifically, we conducted a broad theoretical and experimental study of new methods for generating coherence and explored the new forms of coherence which were found in order to develop technology in areas, where we have unique expertise, in order to solve current problems associated with national security interests. For example, we further developed our FAST CARS techniques and standoff detection schemes based on superradiance to allow for the possibility of detecting bio-chemical agents and explosives in the air via real time stand off spectroscopy. We also explored more efficient methods of generating UV light via quantum coherence.

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Final Report

Contract Number	Grant No. N00014-09-1-0888
Title of Research	Detection of Biochemical Pathogens, Laser Stand-off Spectroscopy, Quantum Coherence, and Many Body Quantum Optics
Principal Investigator	Marlan O. Scully
Organization	Texas A&M University, Department of Physics

Goal:

In the present work, we continued to identify and transition technologies that are of interest to the Navy. Specifically, the objective of the current proposal was a broad theoretical and experimental study of new methods of generation of coherence and exploring new forms of coherence that can be induced in various quantum systems. In particular, we extended approaches based on induced quantum coherence in molecules to challenging problems of great importance for homeland security, e.g., our standoff detection based on superradiance, development of efficient methods of generation of THz, UV and X-rays based on quantum coherence, etc..

As shown in previous work, a major impact of our approach is the ability to acquire signals from infectious agents quickly. For example the optimization of the FAST-CARS setup has yielded single-shot detection of a small amount of *Bacillus subtilis* endospores (10,000 spores). With the pump, Stokes, and probe wavelengths in the near-IR domain, the light-induced damage threshold for the spores was found to be relatively high. It was possible to bring up the pulse energies, reduce acquisition times, and carefully optimize the bandwidth and delay of the probe pulse. As expected, the use of a non-zero, optimal probe delay enhances the contrast between the Raman-resonant and NR contributions. The figure below shows CARS spectra of *B. subtilis* spores taken at the optimized setup in a few laser shots. Spore Raman transitions in the fingerprint region (1300- 1700 cm^{-1}) can be seen even after a single shot. For these measurements, the laser system was switched from normal (continuous) operation at 1 kHz rep rate into the single-shot mode, allowing the laser pulses to be fired manually. The integration time of the CCD camera was set to 3 seconds to ensure a suitable time span for the manual firing of up to ten shots. It should be noted that these data are acquired from back-scattered light off a surface coated with the bacterial spores.

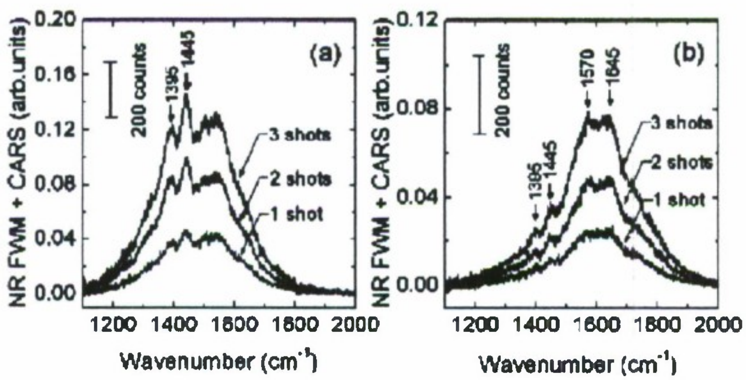

Figure 6: *Bacillus subtilis* CARS spectra for one, two, and three laser shots and the optimal probe delay. Parameters are: pump wavelength $\lambda_1 = 1.25\ \mu$m, 4 μJ/pulse; probe wavelength $\lambda_3 = 805.8$ nm, $\Delta\omega_3 = 30$ cm^{-1}, 3 μJ/pulse. (a) Stokes wavelength $\lambda_2 = 1.54\ \mu$m, 4 μJ/pulse; (b) Stokes wavelength $\lambda_2 = 1.56\ \mu$m, 4 μJ/pulse. (Data from TAMU.)

Overall, the results convey the utility of the approach and its potential for ''on-the-fly'' detection of infectious agents, such as *Bacillus anthracis*.

Accomplishments:

We utilized new understandings of physics and made breakthroughs on the detection of trace gases and biological molecules, extended the limits of CARS spectroscopy, and developed new radiation sources as well as continued investigation into new methods of generation of coherence. We also studied the new forms of atomic coherence found in order to develop technology in areas where we have unique expertise to solve current problems associated with national security interests and needs. A summary of accomplishments follows.

II. Activity

B. Research: Current Activities

We continued research into new approaches to a broad range of problems, for example, detection of biothreats such as anthrax and viral agents via FAST CARS technology as developed in our laboratories and standoff detection based on gain-swept superradiance spectroscopy that will allow us to detect biochemical pathogens and explosives in the air and on the ground via real time stand-off spectroscopy. In addition, we continued investigations into the fundamentals of laser physics, including methods for generating XUV based on quantum coherence, investigations into of cooperative spontaneous emission and the collective Lamb Shift, Electromagnetically Induced Transparency (EIT), and other various fundamental processes. Specific detail of work done is given below.

1. FAST CARS Work and Stand-off Spectroscopy

- Spatial orientation of molecules is a pervasive issue in chemical physics and, by breaking inversion symmetry, has major consequences in nonlinear optics. We developed and analyzed an approach to molecular orientation which extracts from an ensemble of aligned diatomic molecules (equally AB and BA, relative to the E vector) a subensemble that is oriented (mostly AB or BA). Subjecting an aligned molecule to a tailored infrared (IR) laser pulse, we created a pair of coherent wave packets that correlated vibrational phase with the AB or BA orientation. Subsequent, suitably phased ultraviolet (UV) or visible pulses were used to dissociate one of these vibrational wave packets, thereby "weeding out" either AB or BA but leaving intact the other orientation. Molecular orientation has significant implications for coherent Raman spectroscopy. In the absence of orientation, coherence between vibrational levels is generated by a pair of laser pulses off which a probe pulse is scattered to produce a signal. Orientation allows direct one-photon IR excitation to achieve (in principle) maximal Raman coherence.
- We studied the dynamics of a two-level system driven by a strong off-resonant electromagnetic field. We derived an analytical solution for arbitrary pulse shape. We explored possible applications and tested experimentally as appropriate.

- We demonstrated a femtosecond-oscillator-based system for coherent anti-Stokes/Stokes Raman scattering microscopy, wherein impulsive Raman excitation is combined with narrowband, time-delayed, and therefore, background-free probing. We showed that this simple technique could be used for microscopic imaging with chemical contrast.

- We explored the possibilities of using coherent Raman spectroscopy for real-time detection of biohazards. We examined robustness and definitive means for obtaining a molecule-specific signal to be used in species identification by exciting vibrational coherence on more than one Raman transition simultaneously. In particular, we concentrated on detecting dipicolinic acid (DPA), known to be a maker molecule for bacterial spores. We considered time-and frequency- resolved techniques for coherent Raman spectroscopy and adapted these for this particular application.

- We studied interference between a local oscillator and coherent anti-Stokes Raman scattering signal fields by controlling their relative phase and amplitude. This control allowed direct observation of the real and imaginary components of the third-order nonlinear susceptibility of the sample. In addition, we demonstrated that the heterodyne method can be used to amplify the signal.

- We previously demonstrated that by using pairs of pulses of different colors (e.g., red and blue) it is possible to excite a dilute ensemble of molecules such that lasing and/or gain-swept superradiance is realized in a direction toward the observer. This approach was a conceptual step toward spectroscopic probing at a distance, also known as standoff spectroscopy. In the present work, we examined a related but simpler approach on the basis of the backward-directed lasing in optically excited dominant constituents of plain air, N_2 and O_2. This technique relied on the remote generation of a weakly ionized plasma channel through filamentation of an ultra intense femtosecond laser pulse. Subsequent application of an energetic nanosecond pulse or series of pulses boosted the plasma density in the seed channel via avalanche ionization. Depending on the spectral and temporal content of the driving pulses, a transient population inversion was established in either nitrogen- or oxygen-ionized molecules, thus enabling a transient gain for an optical field propagating toward the observer. This technique resulted in the generation of a strong, coherent, counter-propagating optical probe pulse. Such a probe, combined with a wavelength-tunable laser signal(s) propagating in the forward direction, makes possible a tool for various remote-sensing applications. The proposed technique can be enhanced by combining it with the gain-swept excitation approach as well as with beam shaping and adaptive optics techniques.

2. Fundamental Laser Physics

Novel Methods of Radiation Generation and Optical Device Mechanics

- We experimentally demonstrated an ultra-dispersive optical prism made from a coherently driven Rb atomic vapor. The prism possessed spectral angular dispersion that is 6 orders of magnitude higher than that of a prism made of optical glass; such angular dispersion allows one to spatially resolve light beams with different frequencies separated by a few kilohertz. The prism operated near the resonant frequency of atomic vapor and its dispersion was optically controlled by a coherent driving field.
- We studied the selective reflection of the laser beam from rubidium atomic vapor at the D-2 line (wavelength lambda = 780 nm) at different atomic densities. We used a tunable free-running diode laser. We found a measurable signal at a low atomic density N when the mean distance between resonance atoms reached two wavelengths. In our experiment, the dimensionless parameter N (1/3) lambda varied from 0.5 to 2.8. The reflectivity increased with density monotonically.
- We studied the superfluorescence (SF) from a gas of rubidium atoms. The atoms of a dense vapor were excited to the $5D$ state from the $5S$ state by a two-photon process driven by 100-fs laser pulses. The atoms decayed to the $6P$ state and then to the $5S$ state. The SF emission at 420 nm on the $6P$–$5S$ transition is recorded by a streak camera with picosecond time resolution. The time duration of the

generated SF was tens of picoseconds, which was much shorter than the time scale of the usual relaxation processes, including spontaneous emission and atomic coherence dephasing. The dependence of the time delay between the referenced input pulse and SF was measured as a function of laser power. The experimental data were described quantitatively by a simulation based on the semiclassical atom-field interaction theory. The observed change in scaling laws for the peak intensity and delay time could be elucidated by an SF theory in which the sample length was larger than the cooperation length.

- Enhancing nonlinear processes at the nanoscale is a crucial step toward the development of nanophotonics and new spectroscopy techniques. We demonstrate a novel plasmonic structure, called plasmonic nanocavity grating, and found it to dramatically enhance surface nonlinear optical processes. The device consisted of resonant cavities that were periodically arranged to combine local and grating resonances. The four-wave mixing signal generated in our gold nanocavity grating was enhanced by a factor up to 2000, 2 orders of magnitude higher than that previously reported.
- We generated coherent UV light pulses by the coherent scattering of IR pulses from atomic rubidium vapor. Rubidium atoms were first excited by a 100 fs pulse from the 5S ground state to the 5D state via a two-photon transition. The atoms were then pumped by an IR pulse resonant to the 5D–12P transition. The presence of the IR pulse triggered the instantaneous emission of a UV light pulse on the 12P–5S transition. The pulse had a time duration of tens of picoseconds, which was measured by a picosecond-resolution streak camera. The temporal shape of the generated light was explained by a simplified atom–field interaction theory.
- We experimentally and theoretically studied the carrier-envelope-phase (CEP) effects on the population transfer between two bound atomic states interacting with pulses consisting of many cycles. Using intense radio-frequency pulse with Rabi frequency of the order of the atomic transition frequency, we examined the influence of the CEP on the control of phase-dependent multiphoton transitions between the Zeeman sublevels of the ground state of 87Rb. Our scheme had no

limitation on the duration of the pulses. Extending the CEP control to longer pulses created interesting possibilities for generating pulses with an accuracy that was better than the period of optical oscillations.

- We studied the selective reflection from a glass rubidium vapor interface with a large incidence angle in a pump probe experiment. Obtained results indicated that this technique could be used to develop a variable optical filter or optical switcher. Estimates made suggested that the switching time could be less than the natural lifetime of the excited state. A multi-reflection scheme could act to increase the contrast of the variable spectral filter.

<u>Cooperative Spontaneous Emission, Superradiance, and the Lamb Shift</u>

- We studied the evolution of timed symmetric N-atom state prepared by conditional absorption of a single photon and exhibiting superradiant decay. We found analytical expression for the initial decay rate of the state valid for any size of spherical atomic cloud. We showed that the timed symmetric state is only approximately an eigenstate of the system for a large atomic cloud even if virtual processes are neglected.

- We analyzed the collective Lamb shift and associated radiative decay of a large cloud of radius R containing N atoms uniformly excited by one photon of wavelength λ. We showed that the time evolution of the symmetric state prepared by single photon absorption in the limit $R > \lambda$ is similar to that encountered in the Dicke limit of small sample ($R < \lambda$) superradiance. The theory included virtual (counterrotating) terms naturally and thus provided a simple calculation of the collective Lamb shift of a single Dicke state.

- The problem of single photon collective spontaneous emission, a.k.a. superradiance, from N atoms prepared by a single photon pulse of wave vector $\vec{k}_0$ has been the subject of recent interest. It has been shown that a single photon absorbed uniformly by the N atoms will be followed by spontaneous emission in the same direction. Extending this work we have found a new kind of cavity QED in which the atomic cloud acts as a cavity containing the photon.

- We presented analytical solutions for the evolution of collective state of N atoms. We found a (timed) Dicke state prepared by the conditional absorption of a single photon which exhibited superradiant decay. This is in strong contrast to the evolution of a symmetric Dicke state which is trapped for large atomic clouds. We showed that virtual processes yield only a small effect on the evolution of the rapidly decaying timed Dicke state. However, they change the long time dynamics from exponential decay into a power-law behavior which can be observed experimentally.
- We considered collective spontaneous emission from an ensemble of N identical two-level atoms prepared by absorption of a single photon-a.k.a. single photon Dicke superradiance. We found dynamical properties of superradiance for small and large atomic cloud. Moreover, we addressed the effects of virtual processes on collective decay rate and Lamb shift. It turned out that virtual processes lead to relatively small yet interesting effects on the time evolution of a rapidly decaying state. However, such processes substantially modify the dynamics of trapped states by bringing in new channels of decay.

Electromagnetically Induced Transparency (EIT)

- We experimentally studied the propagation of two optical fields in a dense rubidium (Rb) gas in the case when an additional microwave field is coupled to the hyperfine levels of Rb atoms. The Rb energy levels form a close-Lambda three-level system coupled to the optical fields and the microwave field. It has been found that the maximum transmission of the probe field depends on the relative phase between the optical and the microwave fields. We observed both constructive and destructive interferences in electromagnetically induced transparency. A simple theoretical model and a numerical simulation have been developed to explain the observed experimental results.
- We studied a time response of electromagnetically induced transparency (EIT) in a rubidium vapor to a rapid variation of optical phase. We found a very fast growth of the absorption when the phase of the optical field was abruptly changed, followed by a slow return to the level of steady-state absorption. The

recovery time decreased with increasing optical power. A simple theoretical analysis showed that under our experimental conditions the low power limit of the recovery time was determined by the ground relaxation time. In our case it was defined by a time-of-flight of rubidium atoms through laser beam. The obtained value of the ground state relaxation time was in a good agreement with results of direct measurements by 'relaxation in the dark' method. Our technique based on phase dynamics in EIT could be used for investigation of the ground state relaxation and the fast control of EIT.

- We studied electromagnetically induced transparency (EIT) in diatomic cesium molecules in a vapor cell by using tunable diode lasers. We observed a sub-natural resonance in absorption a certain molecular band at different cesium vapor pressures. The width of the EIT resonance showed a linear dependence on a cesium vapor pressure.

Other Various Fundamental Processes

- We studied the dipole-dipole spectral broadening of a resonance line at high atomic densities when the self-broadening dominates. The selective reflection spectrum of a weak probe beam from the interface of the cell window and rubidium vapor was recorded in the presence of a far-detuned pump beam. The excitation due to the pump reduced self-broadening. We found that self-broadening reduction dependence on the pump power was atomic density independent. These results provided experimental evidence for the disordered exciton based theory of self-broadening, and could be useful for the description of the interaction of a strong optical field with a dense resonance medium.

- Stimulated Raman scattering was first discovered by accident, when a cell with nitrobenzene was introduced inside a ruby laser cavity. The first experimental observation of stimulated Raman scattering clearly demonstrated the great potential of this technique to generate new colors of light and to dramatically enhance the efficiency of weak Raman transition. We have recently revisited those two applications in order to develop high-repetition rate discretely tunable picoseconds laser system based on a very efficient Raman amplification and to introduce a novel optical imaging modality based on selective stimulated Raman excitation with the following acoustic imaging of the area most affected by such an excitation.

- We studied the intensity correlations between two orthogonally linearly polarized components of a laser field propagating through a resonant atomic medium. These experiments were performed in a rubidium atomic vapor. We observed that the correlations between the orthogonally polarized components of the laser beam were maximal in the absence of a magnetic field. The magnitude of the correlations depended on the applied magnetic field. The magnitude first decreased and then increased with increasing magnetic field. Minimal correlations and maximal rotation angles were observed at the same magnetic fields. The width of the correlation function was found to be directly proportional to the excited state lifetime and inversely proportional to the Rabi frequency of laser field.
- We obtained an analytic solution beyond adiabatic approximation by transferring the 1D Schrodinger equation into the Ricatti equation. Then we showed that our solution was more accurate than JWKB approximation. The generalizations of the approach to 3D are being investigated and possible applications of obtained solutions are being discussed.
- Motivated by the recent experiment [Sautenkov, V.A.; Rostovtsev, Yu.V.; Scully, M.O. Phys. Rev. A 2005, 72, 065801], we studied the field intensity fluctuations due to interaction between a laser with a finite bandwidth and a dense atomic medium. The intensity-intensity cross-correlation of two orthogonal, circular polarized beams was controlled by the applied external magnetic field. A smooth transition from perfect correlations to anti-correlations (at zero delay time) of the outgoing beams was observed.
- We revisited the concept of time as it appears in quantum, classical, and statistical physics. By combining special relativity and time-independent quantum field theory, we obtained the time-dependent Schrodinger equation, with time, t, and position, r, being regarded as parameters, not operators. We then examined time

as an argument based on the photon wave function in order to show that the time in quantum mechanics is the same as the time in Maxwell's equations. Finally, time was examined from the perspective of statistical time, i.e., time as derived from thermodynamics.

- The dimensional scaling (D-scaling) method first originated from quantum chromodynamics by using the spatial dimension D as an order parameter. It later found many useful applications in chemical physics and other fields. It enables, e.g., the calculation of the energies of the Schrodinger equation with Coulomb potentials without having to solve the partial differential equation (PDE). This is done by imbedding the PDE in a D-dimensional space and by letting D tend to infinity. One can avoid the partial derivatives and then solve instead a reduced-order finite dimensional minimization problem. Nevertheless, mathematical proofs for the D-scaling method remain to be rigorously established. We examined the D-scaling procedures from the variational point of view. Specifically, we showed how the ground state energy of the hydrogen atom model could be calculated by justifying the singular perturbation procedures. In the process, we saw in a more clear and mathematical way how (Herschbach J Chem Phys 85:838, 1986 Sect. II.A) the D-dimensional electron wave function "condenses into a particle".
- We presented an experimental and theoretical study of the carrier-envelope phase effects on population transfer between two bound atomic states interacting with intense ultrashort pulses. Radio frequency pulses were used to transfer population among the ground state hyperfine levels in rubidium atoms. These pulses were only a few cycles in duration and have Rabi frequencies of the order of the carrier frequency. The phase difference between the carrier and the envelope of the pulses had a significant effect on the excitation of atomic coherence and population transfer. We developed a theoretical description of this phenomenon using density matrix equations and are exploring the implications and possible applications of these results.

- We studied the dynamics of a two-level system driven by a strong off-resonant electromagnetic field. We derived an analytical solution for arbitrary pulse shape. We discussed possible applications and made an experimental demonstration of the results obtained.
- The fundamental limit to photovoltaic efficiency is widely thought to be radiative recombination which balances radiative absorption. We showed that it is possible to break detailed balance via quantum coherence, as in the case of lasing without inversion and the photo-Carnot quantum heat engine. This yielded, in principle, a quantum limit to photovoltaic operation which can exceed the classical one, and still remain in complete accord with the laws of thermodynamics.
- The simplest model of the magnetized infinitely thin electron beam was considered in order to investigate the dispersion equation of the induced Smith-Purcell instability. For the grating which took the depth of the grooves to be a small parameter, the dispersion equation of the induced Smith-Purcell instability was obtained. It was found that the condition of the Thompson or the Raman regimes of excitation did not depend on the beam current but depended on the height of the beam above the grating surface. The growth rate of instability in both cases was proportional to the square root of the electron beam current.
- We experimentally and theoretically studied the phase-dependent interference effects in multi-photon excitation under bichromatic radio-frequency (rf) field. Using an intense rf pulse, we examined the interference between the three-photon and one-photon transition between the Zeeman sub-levels of the ground state of 87Rb in order to determine the carrier-envelope phase of the fields even for long pulses.

C. Publications

1. FAST CARS Work and Stand-off Spectroscopy

1. V. Sokolov, K. K. Lehmann, M. O. Scully and D. Herschbach "Orienting molecules via an ir and uv pulse pair: Implications for coherent Raman spectroscopy," Physical Review A **79**, 053805 (2009).

2. Y. V. Rostovtsev, H. Eleuch, A. Svidzinsky, H. Li, V. Sautenkov, and M. O. Scully "Excitation of atomic coherence using off-resonant strong laser pulses," Physical Review A **79**, 063833 (2009).

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2. Fundamental Laser Physics

Novel Methods of Radiation Generation and Optical Devices Mechanics

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Arlington, VA 22203-1995

February 24, 2012

Dear Mike,

Please find attached SF298 and report Final Technical report for Award No. N00014-09-1-0888 entitled "Detection of Biochemical Pathogens, Laser Stand-off Spectroscopy, Quantum Coherence, and Many Body Quantum Optics," a part of the Congressional Quantum Optics Initiative, which is due on April 14, 2012.

Copies have also been sent to ONR REG San Diego-N66018 (140 Sylvester Rd., Bldg. 140, Room 218, San Diego, CA 92106-3521), The Defense Technical Information Center (8725 John J. Kingman Road, Suite 0944, Fort Belvoir, VA 22060-6218), and The Naval Research Laboratory (ATTN Code: 5596, 4555 Overlook Ave. SW, Washington, DC 20375-5320). Please let us know if anything else is needed.

Best regards,

Marlan O. Scully

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