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14. ABSTRACT
The purpose of the proposal was to study of the effects of quantum noise and coherence in quantum optics and bio-physics. We found new ways to create coherent radiation via Dicke superradiance and new kinds of quantum heat engines operating with quantum coherent systems. We also developed new ways to analyze and visualize plane crashes, e.g., the German flight 9525 crash.

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Proposal to Study the Effects of Quantum Noise and Coherence in Statistical and Bio-Physics

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LONG-TERM GOALS

To study of the effects of quantum noise and coherence in statistical and bio-physics.

ACCOMPLISHMENTS

(Note: reference numbers reference the published results found in the publication list)

<u>In Year I:</u>

• We investigated Rabi oscillations produced by adiabatic pulse due to initial atomic coherence [1]. We found this scheme can be useful for achieving generation of coherent light in the backward direction in the quantum amplification by superradiant emission of radiation (QASER) scheme where modulation of the coupling between light and atoms is produced by Rabi oscillations. Initial coherence could be created by sending a short resonant pulse into the medium followed by a long adiabatic pulse, which would lead to light amplification in the backward direction. In conjunction with the time dependent Rabi oscillations, we have developed a new approach to dynamic black hole entropy. In general, the entropy of a black hole may be obtained in two ways: focusing on the radiating atoms or alternatively focusing on the radiated field. By formulating the Unruh acceleration radiation as a straightforward quantum optics problem, obtaining the Bekenstein-Hawking black hole temperature and entropy, i.e., the famous area theory became almost trivial. Even more interesting was the fact that using a simple "laser-like" model yielded the area theorem via a statistical mechanical approach, based on the acceleration radiation emitted by a cloud of in-falling atoms. This approach eliminated the need for introducing the black hole temperature all together. The results of this work were published in year 3, see [34].

• We investigated parametric resonances and found that the combination resonances produced by periodic modulation of parameters can generate substantial gains in new light amplifiers, acronymed QASER (quantum amplification by superradiant emission of radiation). We studied such gains and investigated their parametric resonance spectral patterns [2]. We used the Floquet theory and developed a projection method that can properly capture gains near the primary resonance and subharmonic frequencies for the Mathieu equation and QASER. This work is further extended via statistical entropy analysis we have developed. In particular, we studied the entropy of a single mode laser operating near threshold and compared the results with the statement frequently made in regard to the study of the maser heat engine: "because maser radiation is in a pure state, its entropy is zero." We also calculated the entropy of an ideal Bose Einstein Condensate (BEC) in a box or in a three dimensional trap and found it revealed unusual, previously unrecognized, features of the Canonical Ensemble [3]. For example, we found that for any temperature, the entropy of the Bose gas is equal to the entropy of the excited particles although the entropy of the particles in the ground state is nonzero. This was explained by considering the correlations between the ground state particles and particles in the excited states. These correlations lead to a correlation entropy which is exactly equal to the contribution from the ground state. The correlations themselves arise from the fact that we have a fixed number of particles obeying Bose quantum statistics. We then explored various examples of the correlations between the ground and excited states in Bose gas. Finally, recognizing that a constraint of fixed total number of particles yields correlation between fluctuation of particles in different states in the canonical ensemble, we demonstrated that below the temperature of Bose- Einstein condensation (BEC), the correlation part of the entropy of an ideal Bose gas is cancelled by the ground state contribution. As a result, we found that in the BEC region the thermodynamic properties of the gas in the canonical ensemble can be accurately described in a simplified model which excludes the ground state and assumes no correlation between excited levels [4].

• We investigated electromagnetically induced transparency (EIT) by dynamically coupling a superradiant state with a subradiant state [5]. The superradiant and subradiant states with enhanced and inhibited decay rates acted as the excited and metastable states in EIT, respectively. Their energy difference determined by the distance between the atoms was measured by the EIT spectra, which rendered this method useful in subwavelength metrology. An x-ray extension of this work follows the TAMU/Princeton experiment. Specifically, we designed two different paths for achieving intense sub-femtosecond X-ray sources [6], namely i) via efficient transformation of the picosecond radiation of the X-ray plasma lasers into the trains of subfemtosecond pulses in a resonantly absorbing medium, and ii) via amplification of HHG radiation in the active medium of the X-ray plasma lasers. We demonstrated experimentally that essentially the same technique can be used for realization of both paths. This technique is a modulation of the parameters of the resonant transition (accordingly in absorbing or amplifying medium) produced under the action of sufficiently strong infrared or optical field. We then explored possible experimental realization of the suggested technique. If successful, coherent intense attosecond X-ray pulses could lead to a fast dynamical imaging of the biological macromolecules and other material nanostructures with a unique combination of a record high temporal and spatial resolution.

• We studied tip-enhanced Raman scattering measurements on Au and bulk MoS_2 substrates using a metallic tip functionalized with copper phthalocyanine molecules and demonstrated similar gap-mode enhancement on both substrates [7]. We compared the experimental results with theoretical calculations to confirm the gapmode enhancement on MoS_2. The functionalized tip approach allowed for suppression of background noise.

• We explored the use of tip-enhanced Raman scattering (TERS) to enhance Raman signals from copper phthalocyanine molecules deposited on a bulk MoS_2 substrate by a nanosized gold tip via electromagnetic enhancement [8]. We conducted a comparative study of MoS_2, gold, and SiO_2 substrates. We found that MoS_2 changed relative intensities of molecular vibrations and provided additional gap-mode enhancement of TERS via interaction with the gold tip, suggesting that this method could possibly improve sensitivity and resolution in single molecule detection and imaging on flat substrates.

• We demonstrated the advanced role of computational mechanics and visualization in science and technology through analysis of the Germanwings Flight 9525 crash [9].

• Finally, we developed a method by which the spectral intensity of an ultrafast laser pulse could be accumulated at selected frequencies by a controllable amount [10], investigated the use of femtosecond laserinduced breakdown spectroscopy (LIBS) for plant species differentiation [11], explored an alternative method for the rapid detection of drought stress in plants using femtosecond laser-induced breakdown spectroscopy (LIBS) [12], and invented a Raman spectroscopic technique for high-throughput stress phenotyping of plants [13].

Year 2:

• We calculated the statistical distribution of the photons from a single-atom quantum heat engine [14], which was originally proposed by Scovil and Schulz-DuBois [H. E. D. Scovil and E. O. Schulz-DuBois, Phys. Rev. Lett. 2, 262 (1959)]. In this heat engine model, a single three-level atom was coupled with an optical cavity and is in contact with a hot and a cold heat bath together. We derived a fully quantum laser equation for this heat engine model and obtained the photon number distribution both below and above the lasing threshold. As the hot bath temperature increased, we found that the population was inverted and lasing light came out. However, we noticed that if the hot bath temperature kept increasing, the atomic decay rate was also enhanced, which weakened the lasing gain. As a result, another critical point appeared at a very high temperature of the hot bath, after which the output light became thermal radiation again. To avoid this doublethreshold behavior, we introduced a four-level heat engine model, where the atomic decay rate did not depend on the hot bath temperature. In this case, the lasing threshold was much easier to achieve and the doublethreshold behavior disappeared.

• While vacuum fluctuation of virtual processes are a well-known feature of quantum electrodynamics, their role in phenomena ranging from the microscopic to the cosmic is less generally appreciated. We investigated virtual photons made real by atoms falling into a black hole [15], see Figure 1 in attachments. We show that atoms falling from outside through a cavity into a black hole (BH) emit acceleration radiation which to a distant observer looks much like (but is different from) Hawking BH radiation. In particular, we find the entropy of the acceleration radiation via a simple laser-like analysis. We call this entropy Horizon Brightened Acceleration Radiation (HBAR) entropy to distinguish it from the BH entropy of Bekenstein and Hawking. This analysis also provides a novel insight into the Einstein principle of equivalence between acceleration and gravity.

• Random Optical scattering severely limits the range and sensitivity of detection techniques within/through turbid media, such as biological samples and security-related materials. This past year, we demonstrated a method for enhancing the coupling of light into a highly scattering medium [16]. We also demonstrated that through microscopic engineering of the optical interface, the optical coupling of light into a turbid material can be substantially enhanced [17]. This improved coupling facilitates the enhancement of the Raman scattering signal generated by molecules within the medium. In particular, we found at least two-orders of magnitude more spontaneous Raman scattering from a sample when the pump laser light is focused into a microscopic hole in the surface of the sample. Because this approach enhances both the interaction time and interaction region of the laser light within the material, its use will greatly improve the range and sensitivity of many spectroscopic techniques, including Raman scattering and fluorescence emission detection, inside high scattering environments.

• We also developed a unique technique for generating directional coherent emissions that could be used to create coherent sources in a wide range of frequencies from the extreme ultraviolet (XUV) to the deep infrared [18]. This was achieved without population inversion by pumping a two-level system with a far-detuned, strong optical field that induces the splitting of the two-level system. This technique holds promise for a new method for manipulating the emission frequency simply through intensity-induced atomic modulation which can be scaled to most frequency regimes using various atomic/molecular ensembles and pump energies.

• We explored the use of low noise intensity correlation microscopy in combination with structured illumination to image quantum emitters that exhibit antibunching with a spatial resolution reaching far beyond the Rayleigh limit [19]. This technique should be of interest in microscopy for imaging a variety of samples, including biological ones. We investigated the underlying theory and simulations, demonstrating the highly increased spatial super-resolution, and identified the requirements for an experimental implementation.

• Using Maxwell-Bloch equations, we show how superradiance can lead to amplification and gain at a frequency much larger than the pumping frequency [20]. This remarkable effect was examined in terms of a simple model involving two coupled oscillators, with one of them parametrically driven. We found that this coupled-oscillator model has a hidden parity-time (PT) symmetry for quantum amplification by superradiant emission of radiation (QASER); thus bringing PT symmetry into the realm of parametrically coupled resonators. We also found that the QASER gain results from the broken-PT-symmetry phase. We then quantized the simplified version of the QASER using quantum Langevin equations to better understand how the system is generated from quantum fluctuations.

• Femtosecond adaptive spectroscopic techniques for CARS (FAST CARS) were developed to suppress the noisy nonresonant background and optimize the efficiency of the coherent optical signals [21]. This perspective focuses on the application of these techniques to nanoscale bio-imaging. We explored their advantages and limitations as well as the promising opportunities and challenges of the combined coherence and surface enhancements in surface-enhanced coherent anti-Stokes Raman scattering (SECARS) and tip-enhanced coherent anti-Stokes Raman scattering (TECARS) and the corresponding surface-enhanced FAST CARS techniques. We found that laser pulse shaping of near-field excitations plays an important role in achieving these goals and in increasing the signal enhancement.

• Ultraviolet (UV) irradiation is an effective bacterial inactivation technique with broad applications in environmental disinfection. This past year, we investigated the effects of aluminum nanoparticles (Al NPs) prepared by sonication of aluminum foil on the UVC inactivation of E. coli bacteria and demonstrated a new radiation protection mechanism via plasmonic nanoshielding [22]. Specifically we observed the direct interaction of the bacterial cells with Al NPs and identified the nanoshielding mechanism via UV plasmonic resonance effects observed. This is a substantial step towards developing improved radiation-based bacterial treatments.
• In addition we demonstrated how fluorescent nanodiamond-bacteriophage conjugates can maintain host specificity [23], demonstrated single-shot chemical detection and identification with compressed hyperspectral Raman imaging [24], and demonstrated how two-photon infrared resonance can enhance coherent Raman scattering [25].

Year 3:

• We showed for the first time [26] that it is possible to realize laser beam focusing at the few-photon level in the four-wave-mixing process, and at the same time reduce the quantum uncertainty in width. The reduction in quantum uncertainty derived directly from the strong suppression of local intensity fluctuations. This surprising effect of simultaneous focusing and reduction of width uncertainty was enabled by multi-spatial-mode (MSM) squeezing, and is not possible via any classical optical approach or single-spatial-mode squeezing. Our results show promise for quantum-enhanced imaging and metrology.

• Tip-enhanced Raman scattering (TERS) is a promising optical and analytical technique for chemical imaging and sensing at single molecule resolution. In particular, we examined TERS signals generated by a gap-mode configuration where a silver tip was coupled with a gold substrate and found it can resolve a single-stranded DNA (ssDNA) molecule with a spatial resolution below 1 nm. To demonstrate the proof of subnanometer resolution, we showed direct nucleic acid sequencing using TERS of a phage ssDNA (M13mp18) [27]. We anticipate that this technique can be extended to the high-resolution imaging of various nanostructures as well as the direct sequencing of other important biopolymers including RNA, polysaccharides, and polypeptides.

• We developed a wide-field coherent anti-Stokes Raman scattering microscopy setup based on picosecondlaser- pumped supercontinuum and used it to demonstrate video-rate imaging with chemical specificity [28].

• Raman spectroscopy is a powerful tool for molecular chemical analysis and bioimaging, which shows an astonishing sensitivity when combined with a huge enhancement by the coherence and surface effects. Noble metal nanoparticles have been commonly used for the spontaneous surface-enhanced Raman scattering (SERS) and for the surface-enhanced coherent anti-Stokes Raman scattering (SECARS) spectroscopies, as they provide large enhancement factors via the electromagnetic and chemical mechanisms. Recently, twodimensional (2D) semiconductors, such as monolayer molybdenum disulfide (MoS2), were used for potential SERS applications as cheaper substrates compared to noble metal nanoparticles. However, the coherent enhancement of SECARS on 2D materials has not been previously explored. To this end, we performed experimental SECARS measurements of pyridine–ethanol solutions containing 2D MoS2 nanocrystals [29] and found a giant chemical enhancement factor of 109 over coherent anti-Stokes Raman scattering (CARS).

• Multi-color fluorescent nanodiamonds (FNDs) containing a variety of color centers are promising fluorescent markers for biomedical applications. We conducted a review of the literature and wrote an article [30] which discussed the current trends in FND's development, including comparison to the early development of quantum dots and highlighted some of the latest advances in fabrication, as well as demonstrations of their use in bioimaging and biosensing.

• We performed a detailed theoretical and experimental investigation of supercontinuum generation in largemode- area photonic crystal fibers pumped by a high-energy, high-repetition rate picosecond Nd:YVO4 laser [31], with the goal of using it as the Stokes beam in coherent anti-Stokes Raman scattering setup. We demonstrated reliable and efficient operation of a coherent anti-Stokes Raman spectroscopy and microscopy setup using this supercontinuum source.

• We studied an ideal Bose gas with a fixed number of particles and temperature in a harmonic trap and showed that below the temperature of BEC transition the canonical ensemble correlations can be very accurately taken into account analytically in a simple effective model [32]. This is found to be true because below the temperature of Bose–Einstein condensation (BEC), the correlation part of the entropy of an ideal Bose gas is cancelled by the ground-state contribution. Thus, in the BEC region, the thermodynamic properties of the gas in the canonical ensemble can be described accurately in a simplified model which excludes the ground state and assumes no correlation between excited levels.

• We calculated the entropy of an ideal Bose-Einstein condensate (BEC) in a three-dimensional trap [33] and found unusual, previously unrecognized features of the canonical ensemble. Specifically, we found that, for any temperature, the entropy of the Bose gas is equal to the entropy of the excited particles although the entropy of the particles in the ground state is nonzero. This was explained by considering the correlations between the ground-state particles and particles in the excited states. We found that these correlations lead to a correlation entropy which was exactly equal to the contribution from the ground state. We then examined the results for correlation functions between the ground and excited states in a Bose gas, so as to clarify the role of fluctuations in the system, giving us a better understanding of the sub-Poissonian nature of the ground-state fluctuations.

• Using a combination of quantum optics and general relativity, we showed that the radiation emitted by atoms falling into a black hole looks like, but is different from, Hawking radiation [34]. This analysis also provided insight into the Einstein principle of equivalence between acceleration and gravity.

• Fröhlich discovered the remarkable condensation of polar vibrations into the lowest frequency mode when the system is pumped externally. For a full understanding of the Fröhlich condensate one needs to go beyond the mean field level to describe critical behavior as well as quantum fluctuations. We found that the energy redistribution among vibrational modes with nonlinearity included is essential for realizing the condensate and the phonon-number distribution [35], revealing the transition from quasi-thermal to super-Poissonian statistics with the pump.

• We showed that indirect spin-spin interactions between effective spin-1/2 systems can be realized in two parallel one-dimensional optical lattices loaded with polar molecules and/or Rydberg atoms [36].

• We proposed a paradigm of heat-powered maser in which two-level masers acts as heat-to-work converters [37]. In contrast to textbook knowledge, this system does not require population inversion or coherent driving and hence can operate with a two-level working medium. Therefore, it is a conceptually different type of maser and, more generally, a conceptually different quantum heat machine. Its autonomous character and "free" power source make this machine technologically enticing.

• Understanding the nonlinear properties of water is essential for laser surgery applications, as well as understanding super-continuum generation in water. Unfortunately, the nonlinear properties of water for wavelengths longer than 1064 nm are poorly understood. We extended the application of the Z-scan technique in water to determine its nonlinear refractive index (n2) and nonlinear absorption (?) for wavelengths in the 1150–1400 nm range, where linear absorption is also significant [38].

IMPACT/APPLICATIONS

The above accomplishements and results hold promise for increasing the sensitivity of signal detection and imaging which has significant implications for everything from medical diagnostics (early diagnosis and detection) to national security and defense as well as the development of advanced computational schemes to analyze catostrophic events such as plane crashes.

PUBLICATIONS

<u>Publications Year I:</u>

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HONORS/AWARDS/PRIZES

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