Next Generation Hazard Detection via Ultrafast Stimulated Raman Spectroscopy

Currently, there still exists a need to detect hazardous materials on surfaces and in the aerosolized state at a variety of ranges dependent on the concept of operation. It has long been believed that spectroscopic methods are the most likely candidates to fill this critical gap for soldier protection. Past investigations utilizing standard spectroscopic techniques have produced reasonable results, but have failed to successfully meet the requirements of range, detectivity, and specificity necessary for this mission. The U.S. Army Research Laboratory (ARL) proposes to use its current capability in ultrafast laser technology to investigate femtosecond laser spectroscopy as a means to overcome the current limitations of spontaneous Raman scattering.

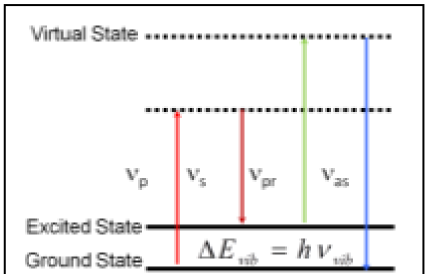

Figure 1: CARS energy level diagram.

The broadband nature of the femtosecond laser pulse has enabled studies involving the coherent and controllable scattering of light (*e.g.,* coherent anti-Stokes Raman scattering [CARS]). The spectral bandwidth of the laser pulse is board enough to coherently and simultaneously excite all the vibrational modes in the molecule of interest allowing for amplification in the anti-Stokes Raman wave (Figure 1). The anti-Stokes radiation is unique to both the molecule and the vibrational mode enabling high chemical specificity and thus aid in the identification of the scattering medium.

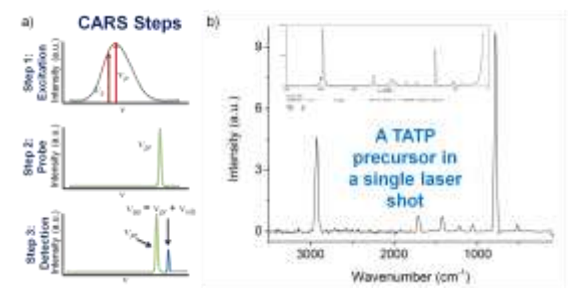

Figure 2: a) Schematic detailing the steps in the multiplex/single-beam CARS experiments. b) Coherent anti-Stokes Raman spectrum of acetone, a precursor to the explosive, triacetone triperoxide (TATP). The spectrum was acquired from a 5 mm cuvette and at an integration time of 1 ms. *Note:* The insert in 2b is representative spectrum from Spectral Database for Organic Compounds (SDBS) obtained using spontaneous Raman.

The research presented here focuses on multiplex-CARS in order to detect threat agents. Single beam or multiplex-CARS avoids the difficult task of maintaining spatial and temporal overlap by combining the pump beam, Stokes beam and probe beam into one, broadband laser pulse. The research performed here demonstrates that CARS scattering has the capability to detect hazardous materials such as dimethyl methyl phosphonate and 2-chloroethyl ethylsulfide (a Sarin and a Mustard chemical warfare stimulant, respectively). Further, evidence shows that CARS is capable of overcoming common sensitivity limitations of spontaneous Raman thus allowing for the detection of material in *milliseconds* as opposed to *seconds*. The exponential increase in scattered photons suggests that the CARS technique may be able to overcome range detection issues found with spontaneous Raman. Further work will investigate limit of detection for hazardous materials at range using surfaces with known roughness and loadings.

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