Remote detection of radioactive materials via Coherent Anti-Stokes Raman Scattering

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Abstract

A sensible nonlinear optical method for remote detection of small quantities of radioactive materials such as deuterium, plutonium, uranium-235, uranium enriched in the isotopes of uranium-233 or uranium-235 with concentration >1 ppb at distances greater than 100 m is proposed. The proposed method is based on remote sensing of the chemo-nuclear by-products (O_3, HNO_3, HO_2NO_2, and NO_2 with the molar fractions ~ $5\ 10^{-7}$, $3\ 10^{-7}$, $2\ 10^{-7}$ and $4\ 10^{-7}$ correspondingly, produced by gamma and neutron radiation interacting with the surrounding atmosphere. The possibility of Coherent Anti-Stokes Raman Scattering (CARS) registration from specific trace gases is discussed.

1. Introduction

The remote detection of Nuclear Materials (NM) is an extremely challenging task. Because the flux of radiation falls as $1/r^2$, standard neutron and gamma radiation detection techniques are typically effective only at ranges of 100 m or less.

A promising alternative for passive remote NM detection is the remote sensing of the chemo-nuclear by-products produced by gamma and neutron radiation interacting with the surrounding atmosphere. Investigators at Los Alamos predicted that ionization of nitrogen by gamma radiation interacting with air will produce a variety of chemical species including nitrogen monoxide, nitrous oxide, nitrogen dioxide and nitric acid [1,2]. The investigators looked for these and other chemo-nuclear species produced by Cobalt-60 radiation, but were limited by the sensitivity of their detectors, 50 ppb.

The remote detection of such trace gases at distances of several meters and at concentrations as low as 1 ppb is now possible using the high-peak-power picosecond-pulse-length lasers. A series of intense picosecond pulses directed at a volume of interest can excite Coherent Anti-Stokes Raman Scattering (CARS) [3,4] from specific trace gases. Moreover, the CARS signal can be remotely amplified using either forward or backward Stimulated Raman Scattering (SRS) from nitrogen. The SRS amplification boosts the CARS signal to a level that allows ppb trace gas concentrations to be detected remotely.

2. Chemo-nuclear trace gas production rates by NM isotopes and background concentrations.

Using a chemical kinetics code, investigators at Los Alamos calculated the expected accumulation of several trace gases as a function of the exposure time to gamma radiation from a 1 Roentgen/s source (113 Ci of ^{60}Co at a range of 20 cm)[i]. The accumulation curves for

- Nitrous Oxide, N_2O;
- Nitric Acid, HNO_3;
- Nitric Oxide, NO; and
- Nitrogen Dioxide, NO_2;

are shown in Figure 1.

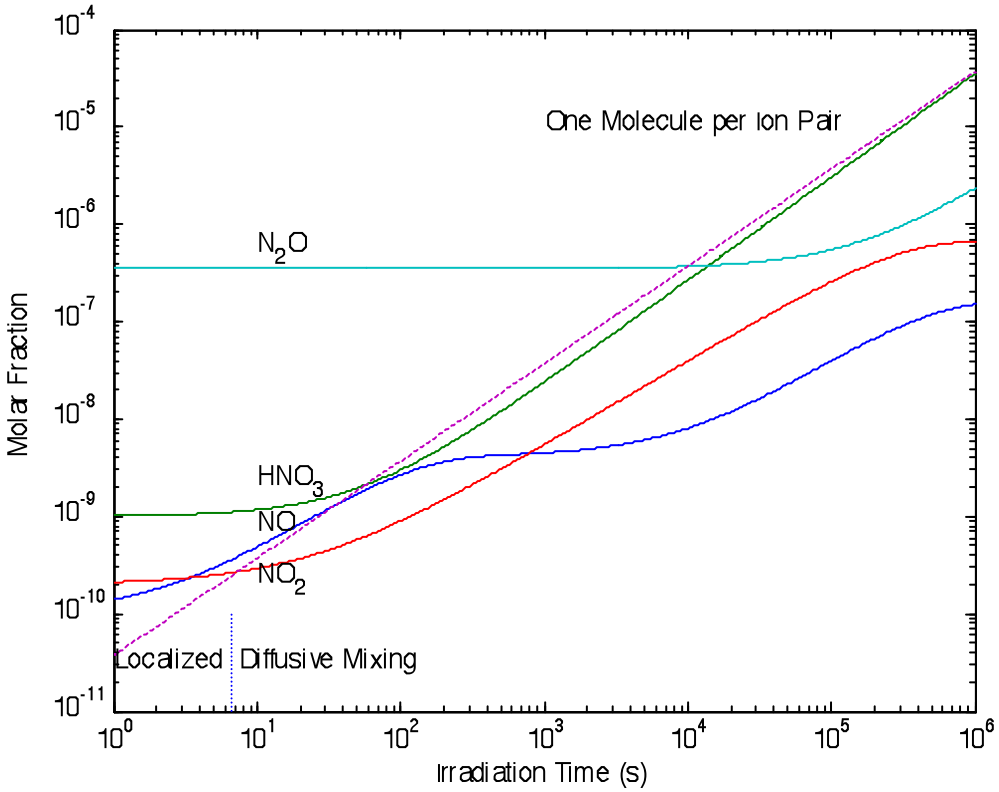

Figure 1: Molar Fraction as a Function of Time for a 1Roentgen/s Source [1,2]

For the most part, the curves are qualitatively consistent with the linear accumulation of trace gases generated by a constant source of ionizing radiation where each ionization event produces gas one molecule. For very short times, the trace gas background concentrations can mask the linear accumulation causing the curves to flatten. For longer times, chemical interactions can deplete the concentrations, again causing the curves to flatten.

For the source 113 Ci of ^{60}Co, at the distance of 20 cm, in still air in the absence of diffusion, several of the trace gases could easily be detected above the background levels after several minutes of irradiation using the amplified CARS technique described below. However, because the activity of the NM isotopes is orders of magnitude lower than the activity of ^{60}Co and because the ionization produced by the keV photons from NM isotopes is substantially lower

than the ionization produced by the MeV ^{60}Co photons, the concentration of trace gases produced by NM isotopes is well below even the ppb sensitivity of remotely amplified CARS. Furthermore, in calculating the accumulation of these gases, the Los Alamos investigators neglected the $1/r^2$ fall off of the gamma radiation field and assumed that diffusion of the trace gases away from the point of production was negligible. In fact, because molecular diffusion velocities in air are typically between 0.1 and 1 $cm/s^{1/2}$, unless the gases are collected in a small sealed container, even in still air diffusion will significantly dilute the concentrations for irradiation times greater than 10 seconds. In open air, this dilution will cause the concentrations to plateau after irradiation times of 10 seconds at or below the ppb level even for the relatively strong gamma source used by the Los Alamos investigators.

In closed containers the diffusion will dilute the trace gases produced by ionizing radiation. Once the container has been shut, the gases will begin to accumulate inside. Over time the concentration will build until the leakage rate of trace gases out of the container is equal to the production rate inside the container. Tests conducted by the German Federal Institute for Materials Research[ii] showed that the exchange rate air in the container with the outside atmosphere can vary from 2% per hour for an unvented container housed in a storage room to 20% per hour for a vented container being transported on the highway. Table 1 shows the expected steady state concentrations of nitric acid produced by different masses of SNM isotopes inside a spherical volume of 33 m^3 (the volume of a standard 20 ft shipping container), at an exchange rate of 2% per hour.

HNO_3	**Source Mass (g)**	
Isotope	**1kg**	**10kg**
U-233	1.54E-10	**1.54E-09**
U-235	4.03E-13	4.03E-12
Pu-239	**1.03E-09**	**1.03E-08**

Table 1: Expected Steady State Concentration of HNO_3 Produced by SNM Gamma Radiation inside a Closed 33 m^2 Container at an Exchange Rate of 2% per Hour

Any gas that leaks from the closed volume will have the same concentration of nitric acid as the air inside the container. Most of the leakage is likely to be concentrated near joint leakage places. At an exchange rate of 2% per hour, the leakage of nitric acid produced by large plutonium and U-233 sources inside a 20 ft container could be detected using the CARS technique. The ppb sensitivity of the system, combined with spatial resolution of the system (~1m) would allow spatial concentration profiles of individual equipment to be mapped.

3. Detecting Chemo-Nuclear Trace Gases with ppb Sensitivity Using CARS

Small concentration of trace gases mixed with other atmospheric gases can be remotely detected by illuminating the target volume with the two picosecond laser pulses of different frequencies. This process is shown schematically in Figure 2. If the frequency difference between the two pulses, $\Delta\omega = \omega_{Pump} - \omega_{Stokes}$, is equal to a Raman vibration frequency of target trace gas, interference between the two pulses will excite Raman vibrations. The vibrations change the gas properties leading to a phonon grating, i.e., a spatial modulation of the index of refraction.

The scattering of the probe pulse from the phonon grating creates a coherent signal at the anti-Stokes frequency, $\omega_{CARS} = \omega_{Pump} - \omega_{Stokes} + \omega_{Probe}$.

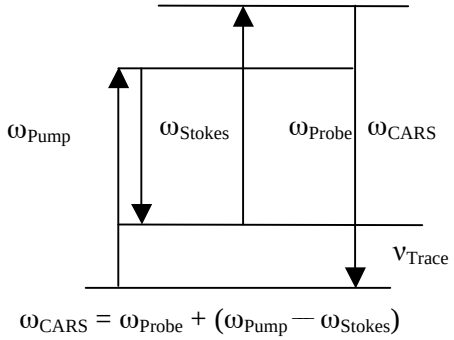

Figure 2: Excitation of CARS

The CARS signal is much stronger than the signal produced by spontaneous Raman scattering. Within a temporal interval of about 100 ps (less than the Raman transition lifetime in air , which is about 140 ps, but longer than the pump pulse widths), a probe pulse of frequency ω_p is directed into the same area. Figure 3 shows the temporal relations between the pulses.

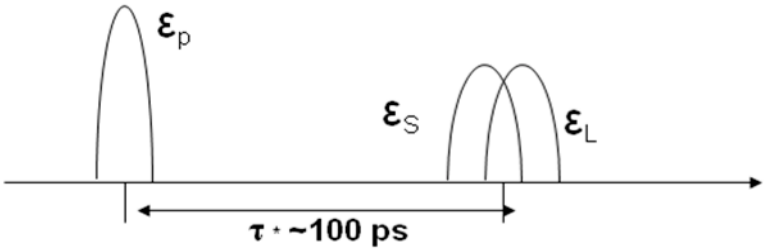

Figure 3: Schema for CARS generation

The phonon amplitude generated within the NO medium, *Q*, is equal to temporal overlap of the pump and Stokes fields such that [3]:

$$Q \sim \int_0^t \varepsilon_S \varepsilon_L dt$$

The phonon grating has a lifetime of some picoseconds.
The energy of the CARS signal will be $E_A = \eta E_P$, where E_P is the probe energy and η is the probe–to–CARS pulse-conversion efficiency, such that:

$$\eta = G_L^2 \times G_S^2$$

G_L and G_A are the total stimulated Raman scattering (SRS) gains. We have $G_{L,A} \sim \sqrt{cE_{L,S}}$ where c is the mixing ratio of the target species while $E_{L,S}$ are the energies of the laser and Stokes emission modes respectively.

For a 100% concentration of the target species at standard temperature and pressure, the gains saturate when the exponent nears a value of 40 (i.e. $e^{40} \sim 10^{17}$) for an input pulse of no more than several millijoules. Thus if a target species mixing ratio of 100 pptv is assumed and this value is inserted into the expression for the gains, the gain values drop to $G_L = G_A = 4 \times 10^{-4}$. Substituting these values into the pulse-conversion efficiency, η, we have $\eta = 2.5 \times 10^{-14}$. So for a probe pulse energy of E_P =10 mJ, the CARS output, will be about $E_A = 2.5 \times 10^{-16}$ mJ, i.e. approximately 300 photons.
In the spectral region near the quadrupled Nd:YAG line of 266 nm, there is virtually no background solar contribution due to absorption by of solar UV by the ozone layer. Any fluorescence in the spectral region is subject both to a slower rise time and lower spectral brightness. The aforementioned 300 photons thus have a high signal to noise ratio and extremely attractive coherence properties.

4. Conclusion

We have discussed the possibility of remote detection of radioactive materials as deuterium, plutonium, uranium-235, uranium enriched in the isotopes of uranium-233 or uranium-235 with concentration >1 ppb at distances greater than 100 m using Coherent Anti-Stokes Raman Scattering in lower atmosphere. We have demonstrated that for a target species mixing ratio of 100 pptv and a probe pulse energy of E_P =10 mJ, the CARS output, will be about $E_A = 2.5 \times 10^{-16}$ mJ, i.e. approximately 300 photons. Such signal can be easily measured using existing methods.

5. References

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[i] http://hallaweb.jlab.org/tech/Detectors/public_html/junk/read/Remote%20sensing%20of%20radiation.pdf