Optimization of ultraviolet Raman spectroscopy for trace explosive checkpoint screening

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Abstract
Raman spectroscopy has long been considered a gold standard for optically based chemical identification, but has not been adopted in non-laboratory operational settings due to limited sensitivity and slow acquisition times. Ultraviolet (UV) Raman spectroscopy has the potential to address these challenges through the reduction of fluorescence from background materials and increased Raman scattering due to the shorter wavelength (relative to visible or near-infrared excitation) and resonant enhancement effects. However, the benefits of UV Raman must be evaluated against specific operational situations: the actual realized fluorescence reduction and Raman enhancement depend on the specific target materials, target morphology, and operational constraints. In this paper, the wavelength trade-space in UV Raman spectroscopy is evaluated for one specific application: checkpoint screening for trace explosive residues. The optimal UV wavelength is evaluated at 244, 266, and 355 nm for realistic trace explosive and explosive-related compound (ERC) residues on common checkpoint materials: we perform semi-empirical analysis that includes the UV penetration depth of common explosive and ERCs, realistic explosive and ERC residue particle sizes, and the fluorescence signal of common checkpoint materials. We find that while generally lower UV wavelength provides superior performance, the benefits may be significantly reduced depending on the specific explosive and substrate. Further, logistical requirements (size, weight, power, and cost) likely limit the adoption of optimal wavelengths.

Keywords Explosives detection · Raman spectroscopy · Cross sections · Trace analysis

Introduction
Standoff trace explosive sensors could potentially improve the security and efficiency of checkpoint screening for a range of venues and events [1–10]. By reducing the role of real-time human interactions and decision-making in the security screening process, standoff trace explosive sensors could increase screening speeds, reduce errors, and decrease costs.

However, this application requires sensing capabilities that can detect limited trace quantities of explosive and explosive-related compound (ERC) residues at nearly real-time speeds and low false alarm rates. The current gold standard for standoff chemical analysis, Raman spectroscopy, can produce chemically specific signatures, but is generally limited to residues with very high mass loads or bulk targets. Inherently, trace chemical sensing (< 100 μg/cm²) presents significant challenges over bulk material detection due to low fill factor, background clutter and substrate interference, and signal variability from particle sizes [10]. These challenges are exacerbated for Raman spectroscopy by two critical limitations: inefficient Raman scattering and interfering fluorescent signals from many substrate and clutter materials.

UV laser excitation has long been suggested as one avenue to push sensitivities and speed relative to visible or near-infrared excitation [8]. Particularly, Raman intensity increases to the fourth power as the excitation wavelength is reduced. In addition, resonance enhancement effects in the UV can increase Raman cross sections by orders of magnitude for many explosives [11]. Finally, if the wavelength is low enough,
background fluorescence interference can be mitigated as the electronic transitions associated with fluorescence emit lower energy light (higher wavelengths) compared with the scattered Raman light.

Despite these benefits, UV Raman has not been adopted for trace chemical sensing applications for several reasons. First, there is a dearth of UV lasers at low enough wavelengths to completely separate fluorescence from Raman signals for most materials. Lasers at < 250 nm which provide adequate output powers do not currently meet size, weight, power, and cost specifications for most operational applications, although this is an active area of research [8]. As a result, most UV Raman sensors must utilize lasers where some fluorescence from background materials must be tolerated. Second, the increase in Raman intensity from shorter UV wavelengths and resonance enhancement is limited by the material penetration depth: the excitation source and the Raman light are both absorbed by many materials. For instance, Emmons and colleagues [12] have shown that Raman intensity of the explosives' ammonium nitrate (AN) and trinitrotoluene (TNT) of different thicknesses can vary greatly with excitation wavelength due to competing effects of UV resonant-enhanced cross sections and shorter light penetration depths.

In this report, we determine the optimum deep-UV wavelength for trace explosive detection on checkpoint-relevant materials. We determine the relative Raman signal using laser excitation at 244, 266, and 355 nm for 8 explosives: AN, composition C-4 (C4), composition B (Comp B), hexamethylene triperoxide diamine (HMTD), potassium chloride (PC), pentaerythritol tetranitrate (PETN), TNT, and urea nitrate (UN). Furthermore, optically thick UV penetration depths are characterized for these explosives via molar absorptivity measurements. As will be seen, total Raman signal intensities for each explosive material vary significantly with excitation wavelength and particle size. The total Raman signals are compared against fluorescent backgrounds from checkpoint-relevant substrates to determine minimum detectable particle sizes. The optimum wavelength for a security checkpoint is determined based on these results. This analysis provides guidance for the critical engineering decision of pump wavelength for security screening and a methodology to conduct the same analysis for other applications.

**Experimental**

**Materials: explosives and explosive-related compounds**

The explosives and explosive-related materials used in this study were obtained from the following sources: The PC, AN, and UN were obtained as solids from Sigma-Aldrich; the 1,3,5-trinitro-1,3,5-triazine (RDX), octahydro-1,3,5,7-
tetranitro-1,3,5,7-tetrazocine (HMX), 2,4,6-trinitrotoluene, and PETN were obtained as analytical standards in solution from Cerilliant through Sigma-Aldrich; the composition B (Comp B), comprised of a mixture of roughly 60% RDX and 40% TNT, and the composition C-4, comprised of 91% RDX and 9% organic binders and plasticizers, were both manufactured by Accurate Energetic Systems, LLC, McEwen, TN, and obtained as a small solid samples through a third party; the Red Dot double-base smokeless powder (SP) was obtained from Alliant Powder; and the hexamethylene triperoxide diamine (HMTD) was synthesized and purified in house, and its purity was confirmed to be > 99% by 1H- and 13C-NMR. Several points should be clarified regarding the materials and their use in these experiments. First, it should be noted that the measurements disclosed include both pure RDX and C4 (~ 91% RDX) for purposes of determining what, if any, differences are caused by the ~ 9% of additives in the C4. All references to these materials in the text and figure captions are correct. Second, the UV absorbance measurements and Raman cross section estimates could not be performed on the SP because it was comprised of a heterogeneous collection of grains, some of which were insoluble in the solvents needed to perform spectrophotometry. Finally, the HMX quantities were limited and experiments requiring “bulk” solid samples could not be performed.

**UV-visible absorption measurements**

UV-visible absorbance spectra were measured for each explosive (AN, RDX, Comp B, HMTD, PC, PETN, TNT, and UN) using a Lambda 1050 Perkin-Elmer dual-beam UV-visible spectrometer. Each explosive material was dissolved in solution at three different concentrations (0.01, 0.005, 0.0025 mg/ml). Comp B, HMTD, HMX, PETN, RDX, and TNT were dissolved in acetonitrile graded for high-performance liquid chromatography (Sigma-Aldrich, ≥ 99.9%) while AN, PC, and UN were dissolved in ultrapure deionized water. Each solution was placed inside of a 1-cm path quartz cuvette. Absorption spectra were acquired between the wavelengths of 200 and 800 nm.

**UV Raman measurements**

Raman spectra for solid-phase explosives and various background substrate materials were measured using laser wavelengths of 244, 266, and 355 nm. Explosive powders (~ 1 mg) were placed in a sample cell with a calcium-fluoride window to allow for > 90% transmittance efficiencies. Background substrates were mounted directly onto the sample holder without a window. A frequency-doubled continuous-wave (CW) argon-ion laser (Lexel Laser 85 SHG) at 10-mW output power was the source for excitation at 244 nm. Two passively Q-switched pulsed microchip lasers (Teem Photonics), with
repetition rates of 2 kHz and pulse-widths of 400 ps, provided light at 266 nm and 355 nm with output powers of 1 mW and 1.8 mW, respectively. For optical efficiency into the slit-based spectrometer, the laser beams were focused onto the sample surface with both cylindrical and spherical lenses, leading to spot sizes of 2.5 mm × 0.1 mm for the beam at 244 nm and 320 μm × 50 μm for the beams at 266 nm and 355 nm. For all wavelengths, samples were irradiated with a laser fluence of 2 J/cm². For fluences above 2 J/cm², photo-bleaching effects were clearly observed on dark substrates and certain explosives are known to photo-degrade [13, 14].

The backscattered Raman light was collected by a 90° off-axis parabolic (OAP) mirror with diameter of 2 in. and focal length of 6 in. An identical OAP mirror imaged the scattered light onto the slit-entrance aperture of the spectrometer with a width of 100 μm. Appropriate long-pass filters (Semrock) were placed before the spectrometer in order to remove the incident laser Rayleigh scattering signal. The spectrometer (Horiba iHR550) utilized a grating with 1200 grooves/mm and a back-illuminated CCD camera (Synapse) with 2048 × 512 pixels. The spectral range of the spectrometers was 100–5000 cm⁻¹ with a resolution of 3 cm⁻¹. The grating was rotated for different pump wavelengths in order to measure the Raman fingerprint region. The charge-coupled device (CCD) camera operated at a readout rate of 20 kHz with a root-mean-square read noise of three counts-per-pixel. Given the collection f-number and CCD pixel size (13.5 μm × 13.5 μm), the depth-of-field was calculated to be approximately 225 μm. All Raman spectral signatures were measured at a 100% areal fill factor, meaning the material filled the entire surface area of the laser beam spot. In addition, all samples were thicker than the depth-of-field of the sensor (225 μm) in order to ensure that Raman signals from transparent UV samples were limited to an approximate interrogation depth equal to the depth-of-field of the spectrometer.

**Results and discussion**

**Molar absorptivity and UV penetration depths**

To determine the penetration depths at UV wavelengths, the absorption spectra of explosives in the solution-phase were measured and converted into the molar absorptivity (also known as the molar extinction coefficient). Using Beer’s law, absorbance values were linearly fit as a function of three different analyte concentrations: 0.01, 0.05, and 0.0025 mg/ml. Molar extinction coefficients were extracted from the slope of the fits. Figure 1a shows the calculated molar absorptivity for nine explosives between 200 and 360 nm. With the exception of HMTD and PC, all of the explosives showed strong absorption below 240 nm, thus contributing to strong resonance enhancement effects as reported by several groups [11, 12, 15, 16].

We define penetration depth as the distance traversed by light into a material until the light intensity is reduced to 1/e (approximately 37%) of its initial intensity due to material absorption. Using the molar absorptivity of the material at the excitation wavelength, ϵ₀(λ) (M⁻¹ cm⁻¹); molar absorptivity at the Raman-scattered wavelength for the most intense Raman cross section mode, ϵ_r(λ) (M⁻¹ cm⁻¹); and the molar concentration, c (mol/cm³), we can calculate the wavelength-dependent penetration depth, L_pen (μm), with the following expression:

$$L_{\text{pen}}(\lambda) = \int_0^D e^{-2.303b[c(\epsilon_0(\lambda)+\epsilon(\lambda))]db}$$

(1)

The integral is evaluated along the laser path length, b (cm), into the material up to the particle diameter, D (μm). We assume that the solution-phase molar absorptivity is a good approximation for the solid-phase molar absorptivity of the explosive. Although there may be some changes to the absorption spectrum due to crystallinity, conformational, and solvent effects, accurately determining penetration depth from solid-state measurements is extremely challenging due to light scattering, so solution-based measurements are the standard approach [12]. When molar absorptivity is minimal, the penetration depth is effectively the particle diameter—meaning the particle is optically transparent and the Raman signal will scale linearly with the particle mass and volume. However, if the material is strongly absorbing, the laser will sample a smaller volume which can significantly reduce the Raman signal compared with an optically transparent particle. Using measured values in Fig. 1a and Eq. 1, we report in Fig. 1b the penetration depths for optically thick explosive particles at common UV Raman laser excitation wavelengths. In the context of Raman spectroscopy, optically thick refers to particle height or material thickness at which Eq. 1 converges to a constant and thus limiting the overall scattered Raman intensity. Penetration depths at 355 nm for all of the materials except Comp B and TNT could not be reported due to very low absorbance values that were below the UV-visible spectrometer noise. These materials were assumed to be UV transparent above 350 nm, and this assumption was validated for one of the explosives, PC, as discussed in the Electronic Supplementary Material (ESM, Fig. S3). More sensitive UV-visible absorption measurements need to be made in order to characterize the penetration depths of the explosives above 300 nm.

Penetration depths vary by several orders of magnitude across wavelength and material. For AN, the results are consistent across all wavelengths with previously reported results [12]. To the authors’ best knowledge, the results shown in Fig. 1b represent the most comprehensive report of explosive penetration depths in the current literature. Comp B, HMX, RDX,
and TNT absorb very strongly between 220 and 250 nm with penetration depths on the order of only 10–100 nm, indicating that the penetration depth will limit the Raman signal for most particles found in trace residues. For instance, at excitation of 244 nm, the optically thick penetration depth for TNT is calculated to be about 15 nm. In this case, the measured UV Raman signal for a thin layer of TNT particles with diameters of 1 μm will be roughly equal to the Raman signal from a thin layer of particles with diameters of 100 μm, assuming the same areal fill factor, even though the analyte mass varies by six orders of magnitude across the two diameters. AN, HMTD, PETN, and UN have much longer UV penetration depths of 1–10 μm, indicating the Raman signal intensity will have a strong dependence on the trace residue particle size distribution.

**Estimated Raman cross section of solid-phase explosives**

The amount of Raman signal, $R_s$, from a material measured on the CCD camera is proportional to the analyte fill factor ($F$) within the laser beam spot size, light collection efficiency ($\eta_c$), spectrometer transmittance efficiency ($\eta_T$), number of excitation photons ($N_{ph}$), analyte molecular concentration ($c$), Raman scattering cross section ($\sigma_R$), and the analyte penetration depth ($L_{pen}$) [17]:

$$R_s = F\eta_c\eta_T N_{ph}c\sigma_R L_{pen}$$

For our Raman measurements, $\eta_c$ of the sensor ($f$-number = 6.4) is calculated to be 0.07% while $\eta_T$ is estimated to be 7%. The overall transmittance-per-wavelength is calculated by the product of the OAP mirror reflectivity, Raman filter transmittance, grating reflectivity, spectrometer mirror reflectivity, and the CCD quantum efficiency. Individual component efficiency values were taken from the manufacturer’s product specifications.

Raman spectra of solid-phase AN, C4, Comp B, HMTD, PC, PETN, SP, TNT, and UN were measured at 100% fill factor at a laser fluence of 2 J/cm² for 244-, 266-, and 355-nm excitation. The solid explosive samples were lightly pressed with a UV transparent window into a sample holder cavity, resulting in sample thicknesses larger than the depth-of-field of the sensor (225 μm). Raman spectra are shown for each explosive and wavelength in Fig. 2. The variations in the spectral intensity across the three excitation wavelengths are driven by both the wavelength-dependent Raman cross section values and penetration depths. We can estimate the solid-phase Raman cross sections in Eq. 2 by integrating the total counts under a specific Raman mode peak, giving $R_s$. We report in Fig. 3 the estimated cross section values for AN, Comp B, HMTD, PC, PETN, SP, TNT, and UN at all three UV wavelengths. Due to significant broadband fluorescence, and lack of strong Raman features, we were unable to determine the cross section for C4 at 355 nm. We were also unable to accurately determine the penetration depth and Raman cross section for SP due to sample heterogeneity. For samples that were UV transparent, the depth-of-field (225 μm) was used as the penetration depth.

The results shown in Fig. 3 should be considered estimates for absolute Raman cross section values. Raman cross sections at 244 nm for AN, C4, and PETN are in reasonable agreement (within a factor of 5) for values reported by Emmons et al. [12]. In order to report an accurate absolute Raman cross section, uncertainty in the optical system and penetration depth must be characterized and reduced. One method involves careful co-deposition of explosive materials, with particles smaller than the...
penetration depth, with an internal standard of known Raman cross section [12]. We note that the cross section values in Fig. 3 do account for the incident and Raman material absorption effects. Furthermore, the system transmission efficiency was verified using the Raman cross section of diamond plates [17]. The close match to previously published results indicates reasonably accurate absolute cross section values. Finally, we note that the solid-phase TNT cross section value at 244 nm is about an order of magnitude lower than published solution-phase measurements [15]. This discrepancy may arise from the different material phases or from sample variability as oxidation of TNT surface can potentially inhibit Raman scattering [18].

We report a factor of 10–15 increase in the Raman cross sections of AN, HMTD, PC, and PETN at 244 nm compared with 355 nm. Similarly, Comp B, TNT, and UN showed an increase of 80–100 at 244 nm. These significant increases in Raman cross section are mostly due to resonant absorption enhancement effects. Pre-resonant enhancement, which increases with a $1/\lambda^4$ dependence, only accounts for a factor of 4.5 increase in Raman signal for a wavelength reduction from 355 to 244 nm.

**Relative Raman signal for trace explosive residues**

Combining Raman cross sections and penetration depth values, we have determined the relative Raman signals across 244, 266, and 355 nm for three different particle diameters (1 μm, 10 μm, and 100 μm) and 8 explosives (Fig. 4). The dimensionless values in Fig. 4 are a product of the last three terms in Eq. 2 (molecular concentration ($b$), cross section ($\sigma_R$), and penetration depth ($L_{pen}$)) and are intrinsic to the material and excitation wavelength. Furthermore, one can multiply the relative Raman signal by the number of incident photons, sensor throughput efficiency, and fill factor to calculate the integrated Raman signal counts for a specific Raman mode.

![Image](image-url)
of an optically thick particle. For materials that are strongly UV absorbing (with small penetration depths), the relative Raman signal follows proportionally to the Raman cross sections and are particle size independent, since the light does not interrogate beyond the minimum particle diameter of 1 μm included in our analysis. For materials that are weaker UV absorbers, the overall Raman signal is greater at 355 nm than at 244 nm for 100-μm-diameter particles due to larger sampling volume, despite lower Raman cross sections at 355 nm. However for realistic fingerprint trace-level residues, particles with diameters of 100 μm are rare; most of the particles will be < 25 μm [18, 19]. In general, our analysis shows that the UV Raman signal for small explosive particles with diameter of 1 μm will be higher at 244 nm compared with 266 nm and 355 nm, assuming the number of photons irradiated onto the particle is fixed. Furthermore, we have incorporated our results of penetration depths with some literature Raman cross section values [12, 15, 20, 21] at other deeper UV and visible/near-IR wavelength to quantify relative Raman signal for trace (1-μm particle diameter)- and bulk (1-mm particle diameter or thickness)-level explosives. We note that these literature cross section values have already been measured and compensated for material absorption at the excitation wavelengths—consistent with our measured Raman cross sections. For trace levels, Raman signal is significantly enhanced in the UV compared with longer wavelength excitation. We conclude that for measuring bulk-level amounts, the optimal wavelength may be in the longer UV or visible range due to longer penetration depths, as long as fluorescence is mitigated. This analysis is reported in the ESM (see Fig. S1).

**Optimal wavelength for trace explosive detection at a security checkpoint**

In any operational setting for trace detection, the Raman signal from explosive particles will also be accompanied by Raman and broadband fluorescence signals from the underlying substrate and environmental clutter materials. We measured Raman spectra of 11 common substrates found at security checkpoints including nylon, leather, plastics, and glass for the three UV bands included in this paper (see Fig. 5). Each spectrum was measured using the same beam size and fluence as the explosive signature measurements. All of the substrate materials exhibit Raman peaks at 244 nm, while many of the materials exhibit broadband fluorescence at 266 nm and 355 nm. Shot noise associated with these interfering signals can ultimately limit the sensor’s ability to detect an explosive particle with high probability of detection ($P_{D}$) and low probability of false alarm ($P_{FA}$). While broadband fluorescence is significantly reduced at 244 nm, overlapping Raman signals from substrate and explosive material can present detection challenges as well.

Fluorescence intensities can be orders of magnitude higher than Raman scattering for materials excited in the visible range, which can be detrimental for trace sensing. However, going to lower wavelengths can effectively separate the Raman fingerprint region from the fluorescing emission spectral window. Beegle and colleagues [22] have shown there is minimal fluorescence emission for some 1000 naturally occurring materials below 270 nm. However, as seen in Fig. 5, certain materials exhibit significantly more fluorescence at
266 nm than at 355 nm. In order to separate the Raman and fluorescence regions entirely, previous reports have shown that wavelengths below 250 nm are desirable [23]. In addition to careful selection of pump wavelength, background fluorescence can be reduced by using time-gated detection because the fluorescence lifetimes of most materials are on the order of few nanoseconds, while Raman scattering occurs nearly instantaneously with laser excitation. For example, one can use a time-gated intensified CCD (ICCD), only detecting low fluorescence signals that occur simultaneously with the Raman signals [24, 25]. In-band Raman interference from the substrate can also be reduced by going to a small laser spot size, effectively exposing less of the background material and thus increasing the threat fill factor. However, this latter approach has several trade-offs, most notably an increase in scan time, and thus cannot provide the sole solution to reducing fluorescence interference.

By combining the measured Raman cross section, penetration depths, and background substrate signals, we can simulate the performance of a notional Raman sensor operating at 244, 266, and 355 nm. Specifically, we determine the minimum detectable explosive particle diameter to achieve a $P_D = 90\%$ and $P_{FA} = 10\%$ on the measured substrate materials across the three excitation wavelengths. The results are shown in Fig. 6. In order to model a sensor optimized for standoff trace detection, the model sensor included in this analysis has a 65× increase in the collection and transmittance efficiencies of the laboratory Raman system used to take signature measurements. Overall light collection efficiency can be increased by reducing the standoff distance, increasing the collection aperture diameter, optimizing optical coatings, and increasing CCD quantum efficiency for the UV. We modeled synthetic Raman spectra of the explosives on the measured substrates at the three UV bands for laser powers of 100 mW and beam diameter of 100 μm on the target. Laser fluence is kept at 2 J/cm². CCD read noise was assumed to be 3.2 counts and dark current to be 1.2 counts/second. Synthetic spectra were calculated by a simple linear mixing model: explosive signal was weighted by the particle areal fill factor within the beam area along with its light penetration depth and the substrate signals were weighted by one minus the fill factor. This model for scaling Raman signals was experimentally validated and reported in the ESM (see Fig. S2). Random Gaussian statistical noise was added to these synthetic spectra to model shot noise and CCD read/dark noise contributions. Alarm decisions were based on the fitting values using linear-least-squares fitting of the synthetic spectra to library threat and clutter spectra.

For excitation at 244 nm, the substrate material type has very little influence on the sensor performance due to minimal interfering fluorescence. The detection algorithm is able to separate the in-band Raman interference from substrate against the threat signatures effectively. For 266-nm excitation, the sensor performance is heavily dependent on material type. Plastics, rubber handles, and certain leather types
fluoresce significantly—thus, $P_D = 90\%$ and $P_{FA} = 10\%$ thresholds are only met at explosive fill factors above 90\%, which minimizes the background signal interference. Certain explosives like C4 and SP at 355 nm are not detectable with these sensor requirements and parameters due to very low threat Raman signals. Additionally, C4 has fluorescence that obscures its Raman features.

We conclude that within our analysis, which includes only excitation at 244, 266, and 355 nm, the optimal wavelength for deep-UV Raman spectroscopy is 244 nm for detecting trace explosive residue particles on common substrates found at security checkpoints due to minimal fluorescence and enhanced Raman cross sections. Our model sensor is easily able to detect explosive fill factors $< 10\%$ for all explosives and substrate combinations at 244 nm. For AN, PC, PETN, and UN, 1\% fill factors are detectable as well. Shorter UV wavelengths may provide improved performance over 244 nm, but a similar analysis of background and threat measurements need to be made in order to quantify any potential enhancements to explosive Raman signals and further fluorescence mitigation. Shorter wavelengths may also reduce the maximum permissible fluence before damage to materials is observed, which can also limit sensor performance.

**Conclusions**

We determined the optimum excitation wavelength between three common UV wavelengths (244, 266, and 355 nm) for Raman spectroscopy of standoff trace explosive detection at a checkpoint. This analysis was completed through comprehensive characterization of both Raman cross sections and penetration depths of eight different explosives as well as the background Raman and fluorescence signals for 11 different common checkpoint substrates. For particle sizes relevant to trace explosive residues, the optimum wavelength is 244 nm. However, due to a lack of compact, economical laser sources at this wavelength, operational deployment of a Raman sensor using 244 nm is currently unlikely, although recent progress has been made to create lasers at wavelengths < 250 nm specifically for tactical Raman spectroscopy (see, for example, The Defense Advanced Research Projects Agency (DARPA) Laser UV Sources for Tactical Efficient Raman (LUSTER) program. [https://www.darpa.mil/program/laser-uv-sources-for-tactical-efficient-raman](https://www.darpa.mil/program/laser-uv-sources-for-tactical-efficient-raman)). In the meantime, commercially available, compact, and economical lasers at both 266 and 355 nm are currently available, but these wavelengths will have limited sensitivity compared with 244 nm. Although 266 nm is generally viewed as more favorable compared with 355 nm due to its lower wavelength and potential resonance enhancement effects, the realized performance may actually be comparable or even worse compared with 355 nm for some operationally relevant explosive/substrate combinations.

Although this paper has focused on wavelength selection for application of Raman spectroscopy for checkpoint screening, there are other considerations as well that determine Raman’s practical utility for this application. The most notable concern is throughput and areal scan rate, and additional analysis is needed to optimize the trade-off between collection optic efficiency and $f/#$, fractional areal coverage, laser power, and spot size, with the resulting probability of detection to identify acceptable concepts of operation. This analysis was, however, beyond the scope of this present work.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflicts of interest.

Research involving human and animal rights This work did not use human subjects or animals in any of the described research.

Disclaimer Opinions, interpretations, conclusions, and recommendations are those of the authors and are not necessarily endorsed by the United States Government.

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