Utilizing Near-Infrared Light Quantum Effects to Measure the Fingerprint Mid-Infrared Region

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Infrared (IR) spectroscopy is a common technique used in both the characterization and identification of molecular compounds. Specifically, the mid-IR fingerprint region (1500 – 600 cm\(^{-1}\)), which is unique for each compound, is utilized in the detection of molecular species. Some applications of this technique include biomedical studies of cancer-related tissues\(^1\), the detection of psychoactive substances in illicit drugs\(^2\), and many more. The potential applications of IR spectroscopy in molecular identification are too numerous to list exhaustively. However, one limitation of this technique has been the high cost of HgCdTe (MCT) detectors.

HgCdTe (MCT) detectors require cryogenic cooling to achieve a good signal-to-noise ratio, which can be rather expensive to maintain over a long period of time. Additionally, these MCT detectors are about 10 to 100 times more expensive than typical silicon (Si) photodiodes, which detect near-IR light with an efficiency 3 orders of magnitude greater at room temperature.\(^3\) Even with cryogenic cooling of MCT detectors, the cheaper Si photodiodes are more efficient with better sensitivity. Therefore, an idea was proposed by Kalashnikov et al. and Mancinelli et al. to use spontaneous parametric down conversion (SPDC) of an incoming photon.\(^4,5\)

Figure 1 illustrates the basic idea behind SPDC in the use of mid-IR spectroscopy.\(^6\) An incoming photon from a pump laser (k\(_p\)) penetrates a finely tuned nonlinear crystal, where most of the photons go straight through while a number of them will split into two non-degenerate photons dubbed signal photons (k\(_s\)) and idler photons (k\(_i\)). As is to be expected, the energy and momenta of both photons is conserved, where the sum of these physical quantities of idler and signaler photons is equal to that of the initial pump photon. Here, the idler photon is of the mid-IR range and comes into direct contact with the material of interest, whereas the signaler photon is in the visible near-IR range. Upon being reflected back through the nonlinear crystal, the pump photons again split into the idler and signal photons. An interference pattern is then detected through differentiation between the first and second generated signal photons, which is based on the idler photon that came into contact with the sample. It is this interference pattern which
is used to generate spectra for the mid-IR fingerprint region. The difference between this setup and the one originally proposed by Kalashnikov et al. in 2016 is the use of the off-axis parabolic mirror, which effectively increases the time idler photons interact with the sample gas. In addition to this mirror, the photon splitting angles ($\theta_s$ and $\theta_i$) are larger to increase the spectral coverage for the mid-IR range, specifically using it for the fingerprint region, whereas previously this was not possible. To demonstrate the application of this method, nitrous oxide ($\text{N}_2\text{O}$) was used, which has absorption in the mid-IR range around 7.69 and 7.88 μm.

Figure 2 graphically shows the interference patterns of $\text{N}_2\text{O}$ at 15 torr (green) and 40 torr (red) as compared to a reference signal in vacuum (dotted line). It is the changes between the vacuum and pressurized patterns that correspond to the IR absorption and refraction for $\text{N}_2\text{O}$. From this, the absorption coefficient is calculated. The change in visibility of the interference pattern, and the refractive index is calculated. These changes are based on the phase shifts of the interference fringes. The interference visibility can be obtained by using the intensity of signal photons observed by the detector. Using this method, both the absorption coefficient and refractive index of $\text{N}_2\text{O}$ were obtained with only a small error or 0.02 cm$^{-1}$ and 10$^{-6}$, respectively.

To conclude, the proposed method here utilizes visible near-IR light to obtain spectral data on the fingerprint mid-IR region via SPDC interferometry. This data is then used to obtain physical data of the gas of interest ($\text{N}_2\text{O}$ in this case) such as the absorption coefficient and refractive index. Using the fact that the fingerprint mid-IR region is unique for each chemical species, the combination of this and the calculated physical quantities can be used in a variety of real-world problems from commercial use to biomedical research.

References:
