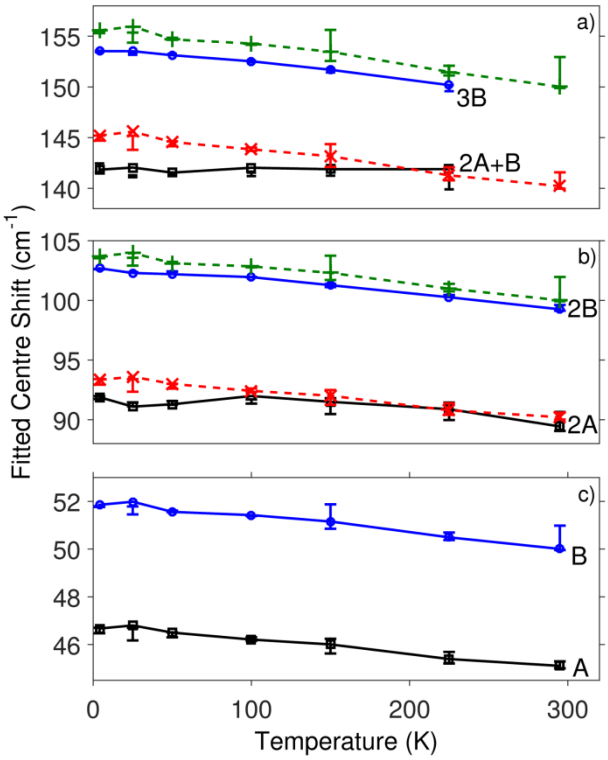
Supplementary Information Document for

Coherence Lifetime broadening of an Optical Transition in a 2 atom diameter HgTe nanowire: a temperature dependent Resonance Raman study.

This section contains details and additional experimental data associated manuscript, and towards the end of this document information on how to utilise the data provided in excel and matlab files.

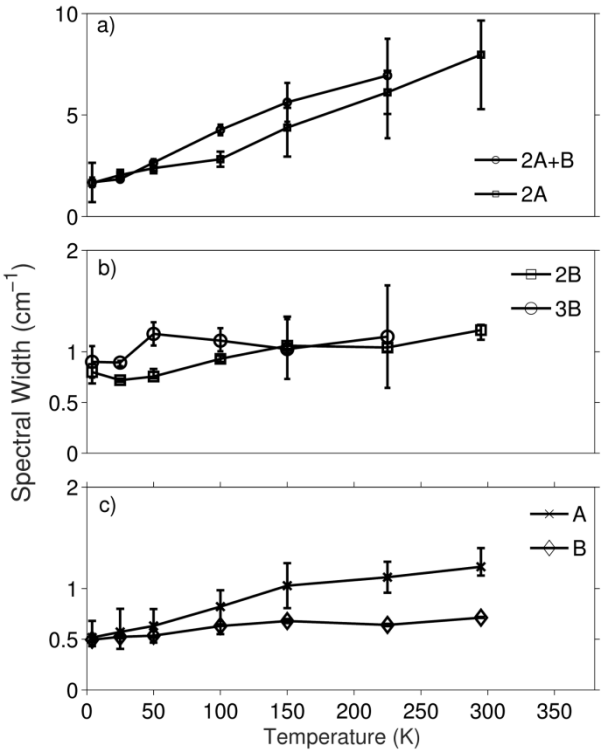
Temperature dependence of phonon modes

Temperature dependent Raman spectra were acquired at the resonance of the B mode at 4K (1.776eV), and the Raman features fit to Lorentzian lineshapes and the peak data extracted. Figure S1 shows the centre shift of the A, B, 2A, 2B, 2A+B and 3B modes as a function of temperature.



S1: Centre shift of Raman modes taken as a function of temperature using incident laser energy of 1.776eV. The dashed lines in panels a and b correspond to multiplications and combinations of the fundamental center shifts from those presented in a. Error bars in all panels correspond to the 95% confidence bounds by fitting Lorentzian lineshapes to the observed modes.

It can be seen by the data presented in figure S1 that as temperature is increased there is a slight softening of the Raman shift for all modes, this is expected and can be understood in terms of lattice expansion.



S2: Fitted spectral width of Raman modes as a function of temperature. The error bars in each panel come from the 95% confidence bands from fitting Lorentzian lineshapes to the modes.

As can be seen from figure S2, in all cases, as the temperature is increased the linewidth of the Raman peaks increases, this can be understood in terms of decrease in the lifetime of the phonon at higher temperatures.

Overall the temperature dependence on the Raman shift and linewidth of the Raman peaks is not too unsurprising.

Fitting of resonances

As reported in the main body of the text multiple models were applied to the observed resonances, to understand the data in terms of sequential, or simultaneous emission. Whilst the main body of the paper reports on the data by applying a Gaussian lineshape, below are the models for double-Lorentzian lineshape for a phonon sequential emission the equation takes the form;

$$L(x) = \frac{A}{((x-c)^2 + b^2)((x-p1-c)^2 + b^2)}$$

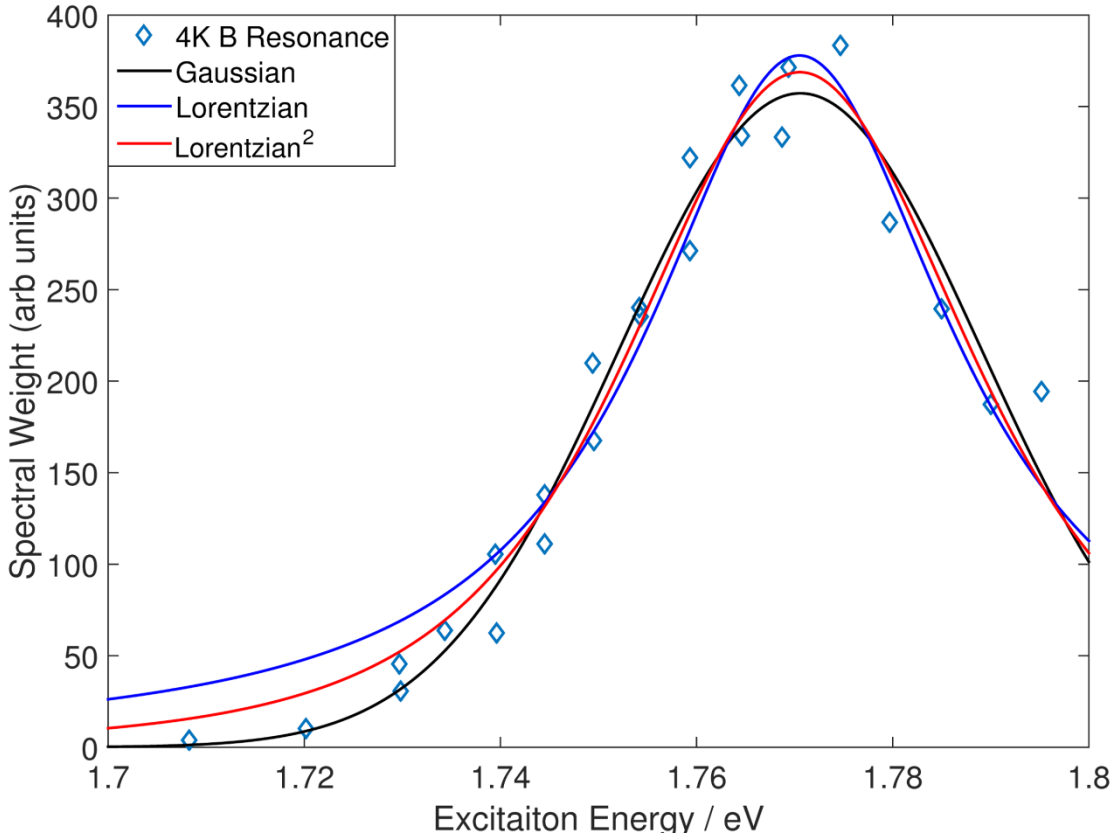
Where c,b and A correspond to fitting parameters for the central energy, linewidth and amplitude and p1 was the measured energy of the fundamental phonon. In the case of 2 phonon sequential emission an extra term of $((x-p2-c)^2 + b^2)$ is added to the denominator, where p2 corresponds to the measured energy of the second phonon emitted.

For an n-phonon simultaneous emission the lineshape takes the form;

$$L(x) = \frac{A}{((x-c)^2 + b^2)((x-pn-c)^2 + b^2)}$$

Where pn corresponds to the measured energy of the phonon involved in the resonance.

As stated in the main body of the text multiple phenomenological fit were applied to the resonances, and there was no clear fit that was superior to the others, for this reason a simple Gaussian lineshape was chosen for the main data. Figure S3 presents the fits applied to the 4K B resonance; the fits include a Gaussian, Lorentzian and double-Lorentzian lineshape. The resulting R^2 value of the fits is presented in tale S1.

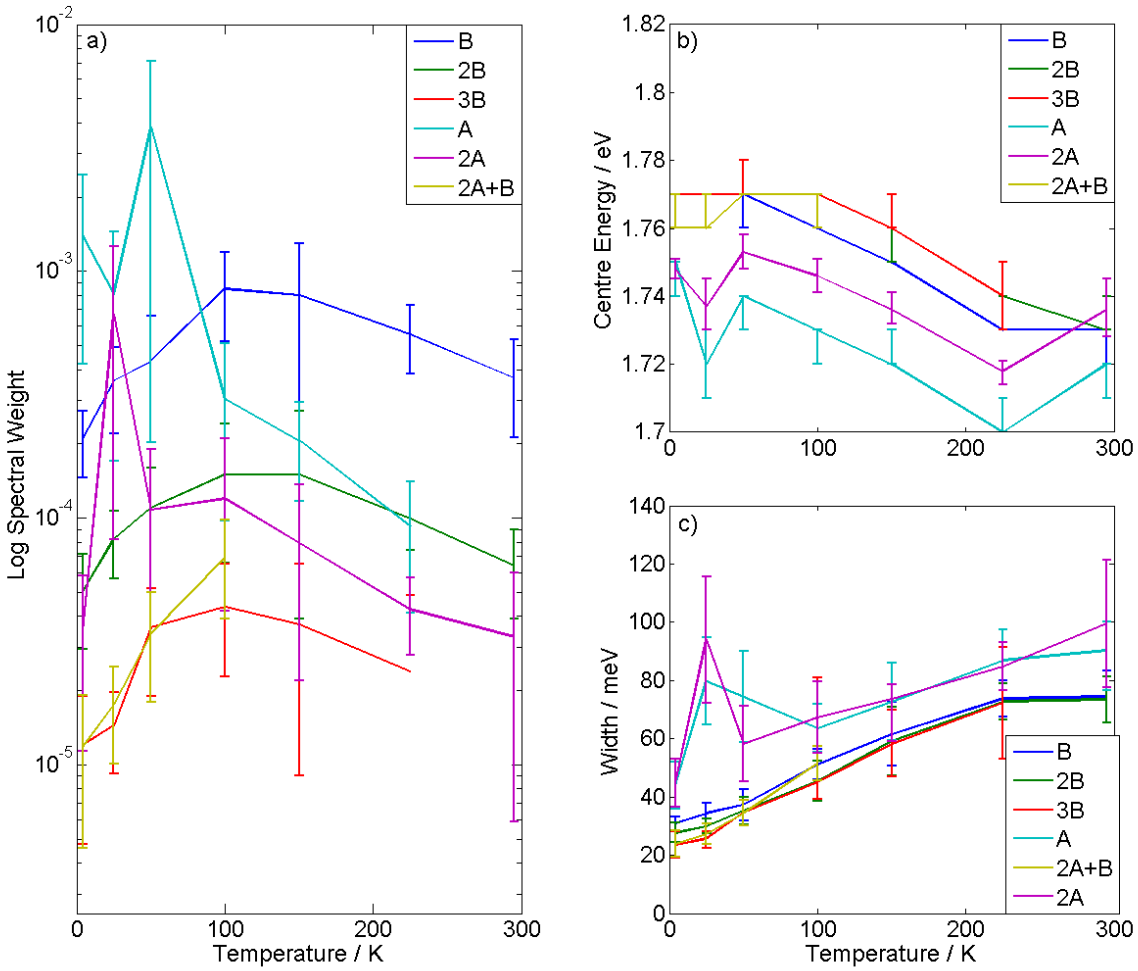


S3: Multiple fits applied to the resonance of the B mode at 4K. Fits include Gaussian, Lorentzian and Lorentzian squared, as it can be seen there is no fit that it clearly superior. The goodness of the fits are presented in table 1.

Lineshape	R^2
Gaussian	0.9692
Lorentzian	0.9516
Lorentzian^2	0.9672

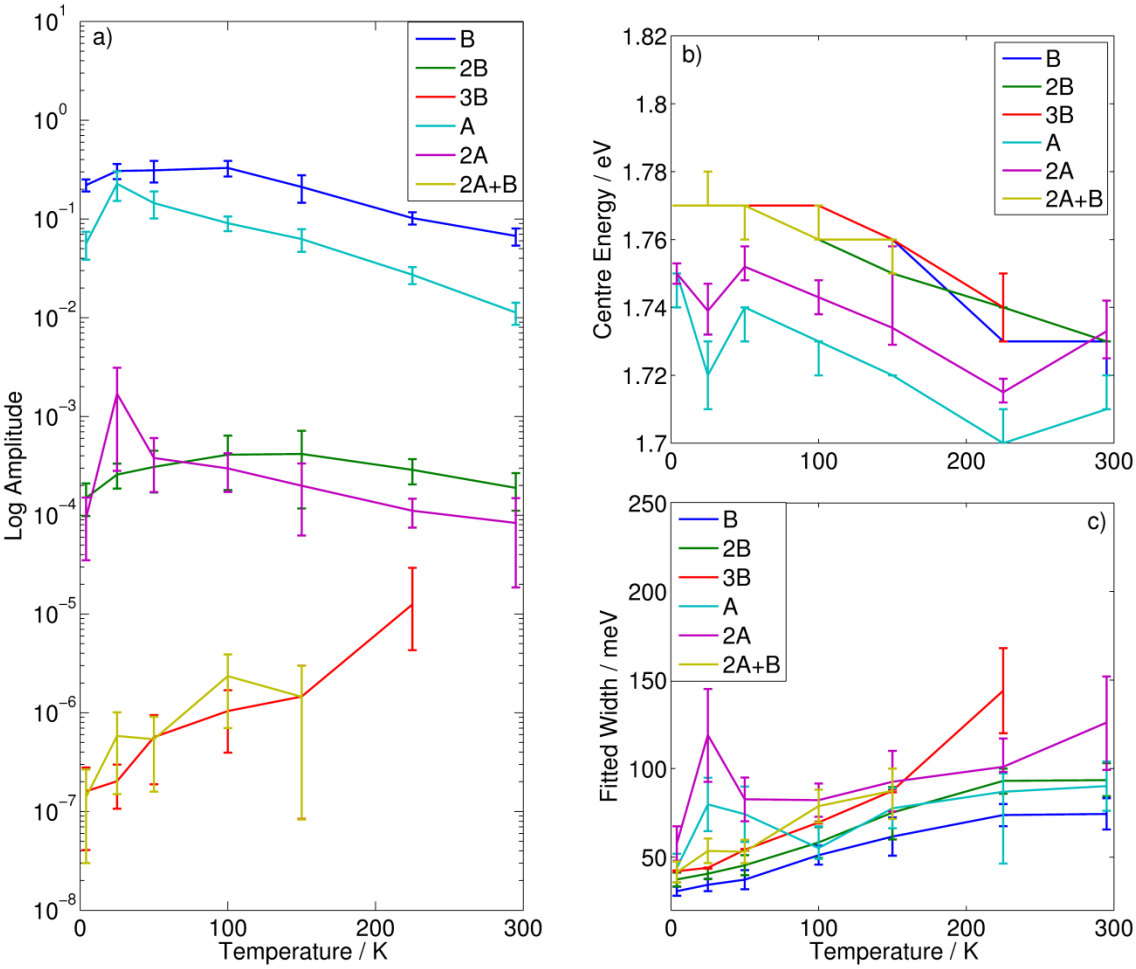
Table S1: R^2 value of fits determined in Matlab for various lineshapes applied, it can be seen that out all the 3 fits, the Gaussian lineshape is marginally the best.

Simultaneous Emission



S4: Extracted parameters of the double Lorentzian fit applied to the A, B, 2A, 2B, 2A+B and 3B modes as a function of temperature for the simultaneous emission model. Panel (a) shows the spectral weight of the resonances, b) the central energy and c) the fitted width. The error bars in each panel are associated with the 95% confidence bounds arising from the fitting.

Sequential Emission



S5: Extracted parameters of the double Lorentzian fit applied to the A, B, a triple Lorentzian to the 2A, 2B and a quadruple Lorentzian to the 2A+B and 3B modes as a function of temperature for the sequential emission model. Panel (a) shows the spectral weight of the resonances, b) the central energy and c) the fitted width. The error bars in each panel are associated with the 95% confidence bounds arising from the fitting.

By inspection of the data presented in figure S4-S5, it is clear from panels (b) and (c) that the central energy and the linewidth of the resonances is has the same behaviour as when fit to a Gaussian lineshape. When considering the amplitude of S4-S5(a) it is fairly clear that there is not a significant change in the amplitude as a function of temperature which suggests that the resonance intensity is controlled by changes in the coherence lifetime as predicted.

Calculation of coherent lifetime

Coherence lifetime of the optical transition was calculated by using the fitted linewidth from the peak data and using equation S1;

$$\tau = \frac{\hbar}{\Gamma} \qquad \text{Eq S1}$$

Comparison to SWCNTs

In the paper by Simon et al referenced in the main text, the authors observe a decrease in the amplitude of the resonance of the inner wall of DWCNTs by a factor of ~10 between 80K and 600K. It is important to note that this is the amplitude of the resonance and not the spectral weight; it was not possible to calculate the spectral weight from the Simon et al. paper because the authors did not provide the linewidths.

The theoretical change in the relative strength of the resonances between 80K and 600K can be calculated by using the predicted linewidths of the resonances as a function of temperature, and accounting for the thermal phonon population.

The value of 1+n for a ~350cm^{-1} RBM of the inner part of a DWCNT was calculated to be 1.002, 1.23 and 1.761 for sample temperatures 80, 300 and 600K. From this it can be shown the the value of 1+n increases by a factor of 1.75 between 80 and 600K.

The width at 80K is ~28meV and at 600K it is ~65meV. These were estimated by inspection of figure 3 from Simon et al paper.

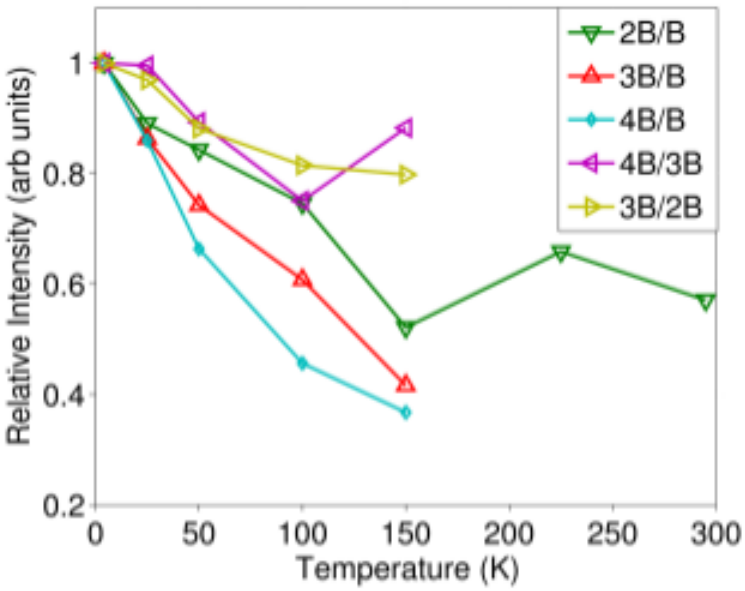
Based on the linewidth dependence it can be predicted that the relative intensity between 80K and 600K should differ by a factor of:

$((65/28)^4)/1.75 \sim 16.5$

Hence we should expect to see a decrease in the relative spectral wright between 80 and 600K by a factor of 16.5 for the inner wall of a DWCNT. When considering the error bars in figure 3 of Simon et al. paper it is reasonable to add a ~30% margin for error on this calculation.

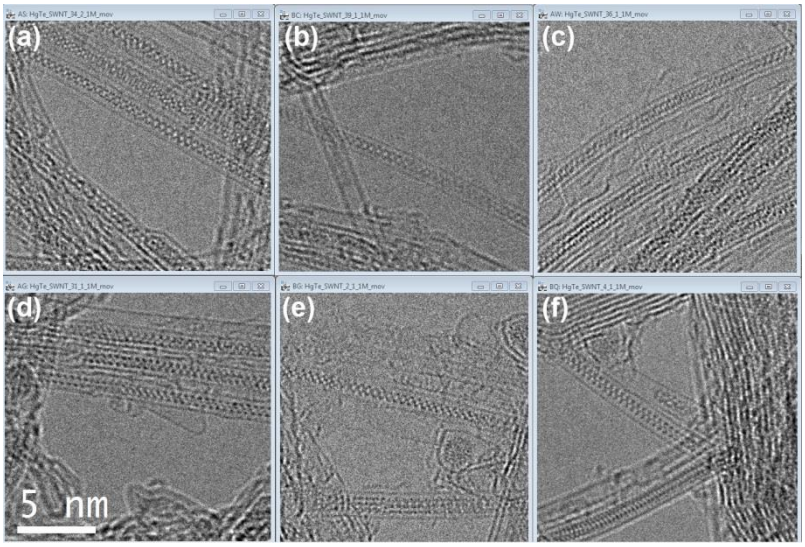
Overall the drop off in signal, whilst not perfect, does not contradict the expected behaviour.

As it is difficult to compare the temperature dependencies of the different peaks on Fig 7 (manuscript), we present the ratios of the fitted amplitudes for various B modes normalized by their 4K value as a function of temperature on Supp Fig 1 (below). It is clear from the data presented on Fig S4 that the ratios are temperature dependent however much less so than the absolute scattering.



S6: Ratios of various fitted harmonics of the B mode as a function of temperature. For clarity, ratios were all normalised by the value at 4K.

The collection of images from a set of filled nanotubes from the Krestinin source presented in figure S7 below show that the filling ratio of the tubular form of HgTe is ~50%.



S7: 6 images associated with the filled HgTe@SWCNTs. Images show that HgTe filling is within at least 50% of the nanotubes sampled.

Supplementary information guideline for data

Provided with this publication includes all the data and fitted parameters obtained from the experiment. This guideline will explain the layout of each file and how the information is presented.

1) The raw data files are provided in MATLAB files labelled as *XK_VBG and ORD Imported Data and Fits.MAT* where X corresponds to the temperature the data was taken at.
   Included in each of these files are the imported spectra for each wavelength. These have 3 distinct file names; intensity_X_Y, shift_X_Y and Wavelength_X_Y, where X and Y correspond to the Temperature and filter type the data was taken at. VBG being the volume bragg grating data, and ORD being the tunable laser line filter data. The only correction for this data is that of the throughput of the spectrometer as discussed in the main body of the manuscript. Additionally in this MATLAB file the Peak data extracted by fitting Lorentzian lineshapes to each mode is also present. This data is also presented in an excel document labelled *'PeakData of Modes'*
2) The Dye laser data for the 3 temperatures are presented in 3 Matlab files labelled 'XK_DyeLaserData' where X corresponds to temperature, like with 1) this data contains the peak data extracted from the fits and the data for each spectra corrected for the throughput. Like with 1) the peak data matrices consist of a number of rows where rows 1,2 are the peak data for the labelled modes, rows 5,6,7,8 corresponds to the width and centre shift of the respective modes.
3) *The excel file 'PeakData of Modes'* as already stated contains the fit parameters for the Lorentzian modes observed. There is a separate info tab in this document to help the user. Each temperature is presented in separate sheets labelled. Within this sheet the first column presents the wavelength, and fitting parameters extracted from the fit. The row of wavelength then shows which wavelength the value associated with each parameter was obtained at. This is then repeated in a block beneath for the confidence bands of the fit and then again for the VBG data. This process again is repeated for the higher harmonics observed.
4) The excel file labelled '*SupplementaryData_EachMode_Shift_Width'* is an excel document which contains the width and shift of the individual modes up to 3B as a function of temperature at a constant excitation energy of 702nm. There is an information box in this document detailing the layout of the data.
5) The excel file *'SupplementaryData_ResonanceProfilesFromFitting'* contains all the fitted parameters from the peak of the resonance by fitting the resonance profile to a Gaussian lineshape. Each block of columns contains the data for each mode labelled A, B 2A 2B etc. Then each column block contains the Amplitude, width and energy of the resonance for each temperature as labelled in the document, also included is the confidence bands from the fit.
6) The excel file *'VBG_ORD_DYE_ModeData_ScalingFactors'* contains the peak data for the A and B modes fitted to Lorentzians as a function of wavelength for 4, 100 and 225K. Within

this spreadsheet at each temperature the scaling factor applied to the B mode to best align the overlap region between the Dye laser with the VBG data and the VBG data with the laser line filter data at each wavelength. The average of this scaling factor was taken and then applied to that relevant temperature.

7) The excel file *FittedResProfileData_SeqSim_AllTemps* contains the fitted resonance data where the data was fit to both the sequential and simultaneous emission model. The document is separated into various labelled tabs at each temperature, where the the final tab summarises all the data in one location.
8) The power dependence data taken to ensure that all spectra are in the linear power regime are given in the data file '*PowerLinearity.xlxs* '. This document contains the extracted peak of the B mode of the HgTe tubes taken at 702nm as a function of incident power at 4K, 50K, and 225K. It was found above 225K the intensity was linear with power up to 2mW, below this temperature non-linear effects began to take place below 1mW, for this reason below 225K the incident power was kept at 0.1mW.