Modelling of nonlinear light up-conversion from intense femtosecond laser pulses

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Abstract. XXX

1. Introduction

The electronic motion associated with chemical reactions or atomic energy transitions takes place on femotsecond to attosecond time scales. Time-resolved imaging of molecular dynamics therefore requires ultrashort laser pulses in the few-femtoseconds regime. A biochemically especially important type of electronic motion is that excited by the absorption of ultraviolet (UV) radiation. A variety of important processes, such as DNA damage, are known to be caused by UV excitation. Generating few-femtosecond UV pulses for time-resolved measurements of UV-excited molecules plays a crucial part in further understanding and eventually controlling these biochemically relevant effects.

The CFEL-ATTO group's STARLIGHT beamline produces few-femtosecond UV pulses by third-harmonic-generation (THG) of a near-infra-red (NIR) input pulse in a noble gas. The THG of an intense ultrashort laser pulse is sensitive to a large range of parameters, including gas pressure, gas density distribution, input beam power, beam focus position, and carrier-envelop-offset (CEO) phase. In order to optimise key properties of the UV output pulse, such as spectrum, energy, and duration, it is therefore advantageous to use theoretical considerations to guide the experimental set-up. This report outlines how, as part of a DESY Summer Student Project, numerical simulations of the THG process were produced using the software package *Luna*. The goal hereby was, in the first instance, to reproduce the experimental conditions of the STARLIGHT beamline and to then carry out various parameter scans in order to find optimal conditions for UV generation. These conditions were then investigated experimentally.

MENTION WHAT HAS BEEN ACHIEVED MENTION THAT FOCUS IN SIMULATIONS ON PARAMETERS THAT CAN BE EXPERIMENTALLY CONTROLLED AT THIS MOMENT (beam power, central pressure, gas)

2. Theoretical Background

This section gives an overview of.... - key properties of ultrashort (few-cycle) laser pulses - the governing propagation equation - relevant nonlinear optical effects/polarisation response

2.1. Nonlinear Optics and THG

For most materials, the polarisation response $\mathbf{P}(t)$ to a moderate electric field $\mathbf{E}(t)$ is linear: $\mathbf{P}(t) = \epsilon_0 \chi \mathbf{E}(t)$, where the linear susceptibility χ is material-specific and generally frequency-dependent. Stronger field amplitudes result in a nonlinear polarisation response, which can be pertubatively expanded in $\mathbf{E}(t)$:

$$\mathbf{P}(t) = \epsilon_0 \left(\chi^{(1)}|\mathbf{E}(t)| + \chi^{(2)}|\mathbf{E}(t)|^2 + \chi^{(3)}|\mathbf{E}(t)|^3 + ... \right) \frac{\mathbf{E}(t)}{|\mathbf{E}(t)|}. \tag{1}$$

In the above, $\chi^{(n)}$ corresponds to the n-th order electric susceptibility. For simplicity, equation (1) leaves the spatial dependence of the polarisation and the driving field implicit. It further treats the $\chi^{(n)}$, which generally are frequency-dependent tensors, as constant scalars. This approximation is valid for isotropic

media such as the noble gases considered here [!!!?? SOURCE]. Note that since gases are centrosymmetric all even-order susceptibilities necessarily vanish, e.g. $\chi^{(2)} = 0$, so that the third-order response becomes the dominant nonlinear term. Due to the small magnitudes of $\chi^{(5)}$ and the other higher-order odd susceptibilities [INCLUDE TABLE OF VALUES!], in the following only the linear and third-order terms of the expansion $\mathbf{P}(\mathbf{E}(t))$ will be considered.

It is evident from (1) how an oscillating electric field in a nonlinear medium results in the generation of co-propagating fields with new frequency components: consider $\mathbf{E}^{(\omega)}(t) = \tilde{\mathbf{E}}\cos(\omega t)$, where all spatial dependence and phase terms have been absorbed into the complex amplitude $\tilde{\mathbf{E}}$. The resulting polarisation response, due to the linear and third-order term, is then given by

$$\mathbf{P}(t) = \epsilon_0 \left(\chi^{(1)} \cos(\omega t)\tilde{\mathbf{E}} + \chi^{(3)} \cos(\omega t)^3 |\tilde{\mathbf{E}}|^2 \tilde{\mathbf{E}} \right) = \epsilon_0 \left(\left[\chi^{(1)} + \frac{3}{4}\chi^{(3)}|\tilde{\mathbf{E}}|^2 \right] \cos(\omega t) + \frac{1}{4}\chi^{(3)}\cos(3\omega t) \right) \tilde{\mathbf{E}}$$
(2)

Thus, the effect of the third-order susceptibility is two-fold: a field contribution $\mathbf{P}^{(3\omega)}(t)$ oscillating at 3ω is generated and an additional term in $\chi^{(3)}|\tilde{\mathbf{E}}|^2$ is added to the polarisation response $\mathbf{P}^{(\omega)}(t)$ at the fundamental frequency. The former process can be identified as THG: a driving electric field oscillating at some frequency will generate a frequency-tripled co-propagating field due to third-order nonlinear interactions in the medium. This is one instance of a range of related nonlinear effects which result in the generation of a higher-frequency field component, known as frequency up-conversion.

Note that a third-harmonic contribution is generated in-phase and co-propagating with the driving wave at each point of the nonlinear medium. Due to dispersion, $k = k(\omega)$, the third-harmonic and the fundamental will generally experience different phase shifts on propagation, resulting in a phase mismatch $\Delta k = k(3\omega) - 3k(\omega)$ between the different third-harmonic components generated at different points along the axis of propagation. This can result in destructive interference of the individual 3ω components. Thus, the phase-mismatch parameter Δk, or, equivalently, the GVD [DEFINE THIS], are important determinants of the achievable THG conversion efficiency η_{THG}, a measure of the energy of the generated third-harmonic relative to the input pulse energy.

The latter process [REWRITE THIS, REFERENCE TOO FAR BACK] is equivalent to an intensity-dependent contribution being added to the refractive index of the medium. The refractive index, n, of a linear medium is commonly defined as $n = \sqrt{1 + \mathbf{P}(t)/[\epsilon_0 \mathbf{E}(t)]} = \sqrt{1 + \chi}$. Analogously, the effective refractive index n_{eff} in a nonlinear medium can be defined as

$$n = \sqrt{1 + \frac{\mathbf{P}^{(\omega)}(t)}{\epsilon_0 \mathbf{E}^{(\omega)}(t)}},$$
(3)

where $\mathbf{P}^{(\omega)}(t)$ is the polarisation response to $\mathbf{E}^{(\omega)}(t)$ oscillating at the fundamental frequency. Due to the presence of the third-order term, and taking into account that $\chi^{(1)} \gg \frac{3}{4}\chi^{(3)}|\tilde{\mathbf{E}}|^2$:

$$n_{\text{eff}} = \sqrt{1 + \chi^{(1)} + \frac{3}{4}\chi^{(3)}|\tilde{\mathbf{E}}|^2} \approx n + \frac{3}{8n}\chi^{(3)}|\tilde{\mathbf{E}}|^2 \equiv n + \Delta n I = n_0 + n_2 I,$$
(4)

where $I = |\tilde{\mathbf{E}}|^2/[2Z_0]$ is the intensity of the driving field, Z_0 the impedance of the wave in the medium, and Δn is the intensity-dependent change in effective refractive index.

This intensity-dependent refractive index has important implications for the propagation of laser pulses in nonlinear media. An important consequence is self-phase modulation (SPM): since the intensity of a beam is significantly higher at its peak than towards the wings, the centre of the beam will experience a higher effective refractive index, and therefore phase-shift on propagation, than off-peak components. This can result in spectral broadening of the pulse [MORE DETAIL???] [IN TIME DOMAIN;]. The intensity-dependent refractive index is also related to effects like self-focussing [SAME THING BUT SPATIAL].

Note that the umbrella term 'Kerr effects' is commonly used to refer to processes associated with $\chi^{(3)}$, including THG, and this practice will be adopted here.

For the present context, there is a further relevant group of nonlinear effects that is highly non-perturbative and therefore cannot be captured by the expansion (1). These are photo-ionisation effects: at sufficiently high field amplitudes, the atoms or molecules in the nonlinear medium become ionised, which affects their interaction with the driving field and generated third-harmonic. Particularly the

values of the $\chi^{(n)}$ and the absorption coefficients are affected. The effects of ionisation on the polarisation response of a quasistatic gas cell are captured by the ADK model. While ionisation limits the achievable third-harmonic pulse energy due to absorption, it affects the spatio-temporal pulse profile and can result in spectral broadening, as is discussed later.

The total polarisation response $\mathbf{P}$ in a nonlinear medium can thus be split up into a linear and non-linear part, $\mathbf{P} = \mathbf{P}^L + \mathbf{P}^{NL}$, where the latter can further be subdivided into Kerr and ionisation effects: $\mathbf{P}^{NL} = \mathbf{P}^{\text{Kerr}} + \mathbf{P}^{\text{ion}}$. MENTION HOW LATER THIS IS IMPLEMENTED IN THE UPPE?

MAKE SURE THAT IT IS CLEAR THAT ALL MATERIALS ARE NONLINEAR AS LONG AS E IS HIGH ENOUGH!!

SAY SOMEWHERE THAT FOCUSSING ON GAS!! BC OF LOW GVD

ALSO MENTION THAT χ IS PRORTIONAL TO MOLAR SUSCEPTIBLITY, HENCE INCREASING DENSITY INCREASES CHI

NOTE: since intensity highest at centre of beam, THG efficiency is more efficient there, so that the THG pulse is narrower, both in space and in time, than the input pulse. Temporally, this is exepcted to by by $\sim \sqrt{3}$ shorter.

USE n0, n2 notation?

SELF-STEEPENING: due to IDRI, peak of pulse experiences higher refractive index and hence (assuming normal dispersion) a slower group velocity than the wings. This results in the leading edge (or trailing edge, for anomalous dispersion) getting steeper. Self-steepening is associated with spectral broadening ?!.

2.2. Ultrashort Laser Pulses

An ultrashort laser pulse can be modelled as a carrier wave at central frequency ω_0 modulated by an envelope function. In the femtosecond regime envelope may only contain a few cycles of the carrier wave.

For simplicity, the following treatment will focus on the temporal and spectral beam profile and neglect spatial contributions. The electric field amplitude of the pulse is then given by

$$\mathcal{E}(t) = \frac{1}{2}\sqrt{I(t)}e^{i[\omega_0 t - \phi(t)]} + \text{ complex conjugate}, \tag{5}$$

where $I(t)$ is the temporal intensity and $\phi(t)$ the temporal phase. It is often more convenient to drop the complex conjugate (analytic signal approximation) and the rapidly oscillating carrier wave phase contribution $e^{i\omega_0 t}$ and work directly with the complex amplitude

$$E(t) = \sqrt{I(t)}e^{-i\phi(t)}. \tag{6}$$

In the frequency-domain, the pulse is described as the Fourier transform of the time-domain field: $\mathcal{E}(\omega) = \mathcal{F}[\mathcal{E}(t)] \equiv \int_{-\infty}^{\infty} \mathcal{E}(t)e^{-iwt}dt$, given by

$$\mathcal{E}(\omega) = \sqrt{S(\omega)}e^{-i\varphi(\omega)}, \tag{7}$$

where $S(\omega)$ is the spectral intensity, or spectrum, and $\varphi(\omega)$ the spectral phase. Since $\mathcal{E}(t)$ is real, the negative frequency regions of $\mathcal{E}(\omega)$ can be neglected.

The temporal and spectral pulse are often expanded as Taylor-series of the form

$$\phi(t) = \phi_0 + t\phi_1 + \frac{1}{2}t^2\phi_2 + ... \tag{8}$$

$$\varphi(\omega) = \varphi_0 + \omega\varphi_1 + \frac{1}{2}\omega^2\varphi_2 + ... \tag{9}$$

where $\phi_n \equiv \frac{\partial^n \phi}{\partial t^n}$ and $\varphi_n \equiv \frac{\partial^n \varphi}{\partial \omega^n}$. It is straight-forward to see from the Fourier transform that the zero-order spectral and temporal phases are the same: $\phi_0 = \varphi_0$. They correspond to what is known as the carrier-envelope-offset (CEO) phase, representing the relative phase between the envelope and the carrier wave. The CEO phase is especially relevant for few-cycle pulses.

The effects of ϕ_1 and φ_1 are spectral and temporal shifts, respectively. Further, ϕ_1 is used to define the instantaneous angular frequency

$$\omega_{\text{inst}}(t) \underset{3}{=} \omega - \phi_1, \tag{10}$$

while φ_1 is used to define the group delay $t_{\text{group}}(\omega) = \varphi_1$.

The term ϕ_2 corresponds to quadratic variations in the temporal phase and hence to a linear variation in the instantaneous frequency with time. This is known as a linear chirp. If the pulse is positively chirped, the frequency increases with time while in a negatively chirped pulse the frequency decreases. Thus, the leading edge of a positively chirped pulse is of a lower frequency than the trailing edge and vice-versa for negatively chirped pulses. In regions of normal dispersion this results in a positively chirped pulse being extended in time and negatively chirped pulses being compressed. The reverse hold in regions of anomalous dispersion.

Thus, nonlinear variations of the temporal phase result in spectral broadening by inducing chirp, which may also affect the pulse duration, if dispersion is present. One such nonlinear variation of the phase in time is given by effects of the intensity-dependent refractive index (IDRI), which results in a larger nonlinear phase increase at the (temporal) centre of the pulse and hence in chirp and spectral broadening if the pulse was initially unchirped. May also lead to spectral compression if the initial pulse was chirped oppositely.

As can be seen from Fourier analysis, the (temporal) pulse length (or pulse width) is inversely related to the spectral bandwidth. Note that the 'pulse length' commonly refers to the full-width-half-maximum (FWHM) length of the pulse, i.e. the time between the most-separated points that are at half of the peak intensity.

———————————————————————— mention dispersion...., GVD,... HERE?

mention how high peak intensities are sufficient in ultrashort pulses to make nonlinear effects relevant

mention bandwith theorem

chirp etc...

GVD

We can expand the dispersion relation $k(\omega)$ around the carrier frequency ω_0:

$$k(\omega) = k(\omega_0) + (\omega - \omega_0)\frac{\partial k}{\partial \omega} + \frac{1}{2}(\omega - \omega_0)^2\frac{\partial^2 k}{\partial \omega^2} + ... \tag{11}$$

The first-order coefficienct $\frac{\partial k}{\partial \omega}$ can be identified as the inverse of the group velocity $v_g = \left(\frac{\partial k}{\partial \omega}\right)^{-1}$ while the second-order coefficient $\frac{\partial^2 k}{\partial \omega^2} = \frac{\partial}{\partial \omega}\frac{1}{v_g}$ is kown as the group velocity dispersion (GVD).

USE k1, k2,.. notation?

GVD IS THE SAME AS BETA2??

We choose $k(\omega) = \frac{n(\omega)}{c}\omega$ (neff??)

The spectral phase acquired by propagating a distance z is: $\phi(\omega; z) = \phi(\omega; 0) + kz \equiv \phi(\omega; 0) + n(\omega)\frac{\omega}{c}z$.

2.3. The Unidirectional Pulse Propagation Equation and Luna

There is a variety of differential equations that can be used to describe the propagation of an ultrashort laser pulse in a waveguide or free space. As is shown in [ref], most of these can be derived from the z-propagated unidirectional pulse propagation equation (UPPE). Several variations exist but its general form for a pulse of frequency ω travelling in the z-direction is

$$\frac{\partial}{\partial z}\mathbf{E}(\omega, \mathbf{k}_\perp, z) = \mathcal{L}(\omega, \mathbf{k}_\perp, z)\mathbf{E}(\omega, \mathbf{k}_\perp, z) + \frac{i\omega}{N_{\text{nl}}}\mathbf{P}_{\text{nl}}(\omega, \mathbf{k}_\perp, z), \tag{12}$$

where $\mathbf{E}(\omega, \mathbf{k}_\perp, z)$ is reciprocal-space electric field amplitude in the frequency domain, $\mathbf{k}_\perp$ is a generalised transverse wavevector, $\mathcal{L}(\omega, \mathbf{k}_\perp, z)$ is a linear operator describing dispersion, diffraction, and absorption, while $\mathbf{P}_{\text{nl}}$ is the nonlinear polarisation with normalisation factor N_{nl}.

The nonlinear polarisation response to a given electric field is computed most easily in real space and the time domain: $\mathbf{P}(t, \mathbf{r}_\perp, z) = \mathcal{P}_{\text{nl}}(\mathbf{E}(t, \mathbf{r}_\perp, z))$, where t is time, $\mathcal{P}_{\text{nl}}$ is an appropriate operator [RELATE TO Pnl ABOVE?!] and $\mathbf{r}_\perp = (r, \theta)$ represents the transverse spatial coordinate. The real-space-time domain representation of $\mathbf{P}_{\text{nl}}$ can then be related to the reciprocal-space-frequency domain version via a temporal Fourier transform, $\mathcal{F}$, as well as an appropriate transverse spatial transform, $\mathcal{T}_\perp$:

$$\mathbf{P}(\omega, \mathbf{k}_\perp, z) = \mathcal{F}\left[\mathcal{T}_\perp\left[\mathbf{P}(t, \mathbf{r}_\perp, z)\right](t, \mathbf{k}_\perp, z)\right]. \tag{13}$$

Note that the real-space-time domain field $\mathbf{E}(t, \mathbf{r}_\perp, z)$ can be recovered from $\mathbf{E}(\omega, \mathbf{k}_\perp, z)$ via the appropriate inverse transformations $\mathcal{F}^{-1}$ and $\mathcal{T}_\perp^{-1}$.

Hence, once the operators $\mathcal{L}$ and $\mathcal{P}_{\rm nl}$ as well as appropriate definitions of the transverse (reciprocal) space $\mathbf{k}_\perp$ and associated transform $\mathcal{T}_\perp$ are known, the UPPE can be solved as an initial value problem in z, using appropriate cycles of transforms at each step.

The choice of $\mathbf{k}_\perp$ and $\mathcal{T}_\perp$ is mostly a matter of the modal expansion of the field in terms of a set of orthogonal spatial modes [the choice of the transform also affects the shape of the linear operator; Note: UPPE derived from Maxwell's equation by substituting a modal expansion!]. These may be waveguide modes or, in the present case, free space modes. The frequency-domain field can be expanded as

$$\mathbf{E}(\omega, \mathbf{r}_\perp, z) = \sum_j \tilde{E}_j(\omega, z)\hat{\mathbf{e}}_j(\mathbf{r}_\perp, z), \tag{14}$$

where $\hat{\mathbf{e}}_j(\mathbf{r}_\perp, z)$ is the orthonormal transverse field distribution of the jth mode and $\tilde{E}_j(\omega, z)$ is the frequency-domain field in mode j. The modes are normalised via $N_{j,\rm nl}$ to ensure that $|\tilde{E}(\omega, z)_j|^2$ represents the spectral power density in the jth mode.

Given an input field $\mathbf{E}(t, \mathbf{r}_\perp, z)$ the modal decomposition can be calculated via

$$\tilde{E}_j(\omega, z) = \mathcal{F}\left[\int_S \hat{\mathbf{e}}_j(\mathbf{r}_\perp, z) \cdot \mathbf{E}(t, \mathbf{r}_\perp, z) d^2\mathbf{r}_\perp\right], \tag{15}$$

where S is the cross-sectional area of the gas cell. Modal decomposition can be used to solve a scalar version of the UPPE for each $\tilde{E}_j(\omega, z)$:

$$\frac{\partial}{\partial z}\tilde{E}_j(\omega, z) = \mathcal{L}_j(\omega, z)\tilde{E}_j(\omega, z) + i\frac{\omega}{N_{j,\rm nl}}\tilde{P}_{j,\rm nl}, \tag{16}$$

where $\tilde{P}_{j,\rm nl}$ represents the projection of the polarisation onto the jth mode:

$$\tilde{P}_{j,\rm nl} = \int_S \hat{\mathbf{e}}_j(\mathbf{r}_\perp, z) \cdot \mathbf{P}_{\rm nl}(\omega, \mathbf{r}_\perp, z) d^2\mathbf{r}_\perp. \tag{17}$$

The linear operator acting on the jth mode can be described as

$$\mathcal{L}_j(\omega, z) = i\left(\frac{\omega}{c}n_{\rm eff}(\omega, z) - \frac{\omega}{v}\right), \tag{18}$$

where $n_{\rm eff}$ is the complex effective index (whose real part represents the effective refractive index [not taking into account IDRI?!] and whose imaginary part gives the attenuation coefficient), c the speed of light in vacuum, and v is the velocity of an arbitrary reference frame. Here, v is taken to be the group velocity at the central wavelength λ_0.

For the present case of pulse propagation in free-space ('free-space' as opposed to in waveguide or fibre; does propagate through gas, not vacuum) the appropriate reciprocal transform $\mathcal{T}_\perp$ is the (zeroth-order) Hankel transform, defined by

$$\mathcal{H} \equiv \int_0^\infty dr J_0(kr)r \tag{19}$$

, where J_0 is the zeroth-order Bessel function. DEFINE THIS IN APPLICATION TO E OR P. Thus, $\mathbf{r}_\perp = (r, \theta)$ and $\mathbf{k}_\perp = (k, \theta)$ where any theta-dependence can be dropped due to the cylindrical symmetry. ... Note that this assumes radial symmetry about the propagation axis, which is justified due to the relevant gemoetry of the gas cell. THUS, ONE-DIMENSIONAL TRANSFORM ONLY IN r?!

The total nonlinear polarisation can be calculated from the total electric field while separating Kerr and ionisation contributions (ignoring higher-order terms of (1)):

$$\mathbf{P_{nl}}(t, \mathbf{r}_\perp, z) = \mathbf{P}_{\rm Kerr}(\mathbf{E}) + \mathbf{P}_{\rm ion}(\mathbf{E}). \tag{20}$$

Assuming that $\chi^{(3)}$ is not dependent of frequency, and hence that THG is instantaneous, as well as approximating the third-order susceptibility as a scalar due to the isotropy of the gas, the Kerr term can be written as

$$\mathbf{P}_{\rm Kerr}(t, \mathbf{r}_\perp, z) = \epsilon_0 \chi^{(3)}|\mathbf{E}(t, \mathbf{r}_\perp, z)|^2\mathbf{E}(t, \mathbf{r}_\perp, z). \tag{21}$$

At sufficiently high intensities, photoionisation effects set in. This affects pulse propagation as the freed electrons are polarisable and as energy is lost to the ionisation potential. [NOTE; the following two things should be n_0 and n_e] If a_0 is the original number of neutral atoms and $a_e(t, \mathbf{r}_\perp, z)$ the number of free electrons at a given point in time and space then a_e can be calculated via

$$\frac{\partial}{\partial t} a_e = (a_0 - a_e) W(\mathbf{E}) \tag{22}$$

where $W(\mathbf{E})$ is the ionisation rate. The ionisation rate can be found via THE ADK MODEL?? [link papers]. Given a_e, the induced nonlinear polarisation due to ionisation can be shown to be

$$\frac{\partial}{\partial t} \mathbf{P}_{\text{Ion}}(t, \mathbf{r}_\perp, z) = \mathbf{E}(t, \mathbf{r}_\perp, z) \frac{I_p}{|\mathbf{E}(t, \mathbf{r}_\perp, z)|^2} \frac{\partial}{\partial t} a_e(t, \mathbf{r}_\perp, z) + \frac{e^2}{m_e^2} \int_{-\infty}^{t} a_e(t', \mathbf{r}_\perp, z) \mathbf{E}(t', \mathbf{r}_\perp, z) dt', \tag{23}$$

where I_p is the ionisation energy, e and m_e are the electronic charge and mass. ———————————
———————————————————————————————————-
———————————————————————————————— NOTE: small e should be changed to mathcal E or something to avoid confusion! not basis vector but field! orthogonal with respect to int dr, not dot product! $\int_S \hat{\mathbf{e}}_j \cdot \hat{\mathbf{e}}_k d^2 \mathbf{r}_\perp = \delta_{jk}$

DUE TO CYLINDRICAL SYMMETRY: expanding on Bessel function basis, not plane waves? Apart from that, free-space transverse modes?

I GUESS CYLINDRICAL WAVEGUIDE MODES? BESSEL FUNCTIONS... -¿ free space modes?!

IMPORTANCE OF SPATIAL TRANSFORM: comes in during UPPE derivatioin?

THERE MIGHT ACTUALLY NOT BE A RELATION BETWEEN MODAL EXPANSION AND HANKEL TRANSFORM??

RELATE UPPE TO MAXWELL'S EQUATIONS! -¿ UPPE CAN BE DERIVED FROM MAXWELL based on a modal expansion -¿ thus slightly different forms for different contexts [link paper]

MENTION LUNA!!!

WRITE ABOUT Pkerr, Pion ,....

MENTION HANKEL TRANSFORM! -¿ find out more! which order zeroth order?? which spatial modes does this correspond to? bessel? -¿ one dimensional Hankel transform essentially?

HOW TO FIND THE SPATIAL MODE VECTORS??? MAYBE VECTORS NOT REALLY SPATIAL!??

POTENTIALLY START WITH THIS SECTION?

Hankel transofmr is used instead of eq 9 for modal decomposition! Hankel transform essentially 2D Fourier transform for radially symmetric function

STILL PLANE WAVES!!

3. The STARLIGHT Beamline
NOTE THAT WE CAN ASSUME LINEARLY POLARISED BEAM

4. Simulation Results
The simulations were carried out with *Luna*, as described above. The simulation parameters were set up to match the experimental parameters are closely as possible.

The gas cell was modelled as a 3mm long radially symmetric chamber with a 0.5mm diameter. The spatial profile of the input beam was taken to be a Gaussian with a $65\mu m$ waist size focussed at the centre of the cell, based on measurements of the spatial beam profile. The input NIR pulse was fed in based on spectral measurements (FROG) of its temporal profile [SENTENCE MAKES NO SENSE]. The central wavelength was taken to be 730nm, based on NIR input pulse measurements, whereas the pulse length of the input pulse was 5.9fs. The pressure at the cell edges was taken to be 1mbar. The total wavelength window considered ranged from 100nm to 1000nm, with the range 100-360nm considered as the UV component and 600-1000nm as the IR component. The nonlinear effects considered where the Kerr effect as well as ADK ionisation, although the latter was turned off in some cases, in order to isolate the effects of ionisation. The CEO phase of the input beam was set to zero without measurement, as simulations showed little impact of the CEO phase on the simulation output, since the input pulse involves sufficient cycles.

Note that two different models for the gas density profile were considered. The first one considered only the cell itself, with a simple density gradient constructed based on specifying the central pressure as well as the edge pressure. The second model was more sophisticated and based on simulations produced with the COMSOL Multiphysics software. It took into account not only the gas cell itself but also the geometry of the 7mm glass chip surrounding it, as well as the effects of the differential pumping system.

The simulations were focussed on investigating parameters that could be experimentally controlled. These were the gas, the central gas pressure, and the input beam power. The gases considered were Argon, Neon, Helium, Krypton, Xenon, Nitrogen, and Nitrous Oxide (N_2O). The beam powers considered were 75mW, 150mW, 200mW, 300mW, and 400mW. The simulated central pressures ranged from 0.1bar to 5.1 bar in 0.1bar increments.

The simulation output included the UV spectrum at the end of the cell (or chip), the total output UV energy, the THG conversion efficiency, the pulse duration of the UV pulse, and the cell (or chip) position at which the UV energy intensity peaked. Additionally, the option to visualise the results in more detail was included, with plots such as: time-domain IR pulse profile, time-domain UV pulse profile, spatiotemporal UV beam evolution, spatiotemporal IR beam evolution, on-axis frequency evolution, UV spectral evolution, on- and off-axis UV and IR intensity profiles, UV and IR energy profiles along z, various spectra.

5. Experimental Results
6. Discussion
Comparison with results at times difficult: pressure and energy of output beam hard to measure

Many more factors to be investigated (beam focus position, beam size, higher beam powers...)

Improve simulations by improving COMSOL gas density model (need to use high Mach number model instead, current model does not take into account gas interaction).

7. Conclusion
Acknowledgments
[1]

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