Vacuum Ultraviolet Generation via Wave-Mixing

Nonlinear medium
- crystals
- metal vapors
- noble gases

Again Maxwell's framework:
coupled wave equations

$$\frac{d}{dz}\mathbf{E}_q = i\frac{2\pi(q\omega)^2}{ck_q}\mathbf{P}_q^{NL}$$

$\mathbf{E}_q$ and $\mathbf{P}_q^{NL}$

Fourier components of fields at frequency $q\omega$
Fields:

$$\mathbf{E}_q = \hat{\mathbf{E}}_q(z,t)\exp[ik_q z]$$

General issues in
four wave mixing or third harmonic generation:

Susceptibility of the medium :
involves transition dipoles in atoms and resonances

Transparency : problems at short wavelengths

Phase-matching :
including the effects of focused laser beams

Practical limitation: optical breakdown

Nonlinear polarization has several terms:

$$\mathbf{P}_3^{NL} = \frac{N}{4}\Big[\underbrace{3\chi_T^{(3)}(3\omega)\mathbf{E}_1\mathbf{E}_1\mathbf{E}_1}_{\text{THG}} + \chi_S^{(3)}(3\omega)\mathbf{E}_3|\mathbf{E}_3|^2 + \chi_S^{(3)}(\omega,3\omega)\mathbf{E}_3|\mathbf{E}_1|^2\Big]$$

Phase-matching and focusing

Three processes of four-wave mixing:

I $\omega_1 + \omega_2 + \omega_3 \rightarrow \omega_4$

II $\omega_1 + \omega_2 - \omega_3 \rightarrow \omega_4$

III $\omega_1 - \omega_2 - \omega_3 \rightarrow \omega_4$

Electric fields:

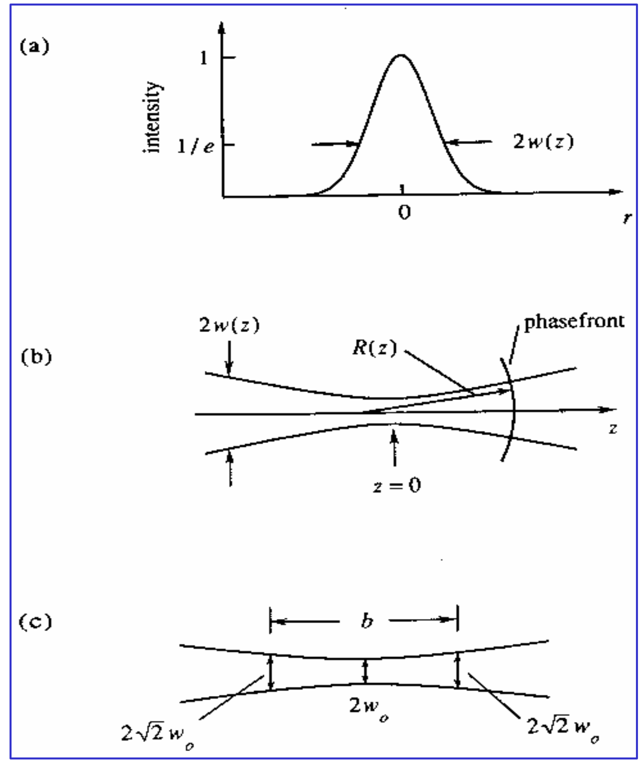
$$\mathbf{E}(\mathbf{r},t) = \mathrm{Re}[\mathbf{E}_1(\mathbf{r})\exp(-i\omega_1 t) + \mathbf{E}_2(\mathbf{r})\exp(-i\omega_2 t) + \mathbf{E}_3(\mathbf{r})\exp(-i\omega_3 t)]$$

Define optical beams as lowest order Gaussians
for each frequency ω
Gaussian beam TM_{00}
(→ higher order modes give differing results !)

$$\mathbf{E}_n(\mathbf{r}) = \mathbf{E}_{n0}(\mathbf{r})\frac{\exp(ik_n z)}{1+i\xi}\exp\left[\frac{-k_n\left(x^2+y^2\right)}{b(1+i\xi)}\right]$$

Confocal parameter b

$$b = \frac{2\pi w_0^2}{\lambda} = \frac{2\pi w_0^2 n}{\lambda_0} = \frac{2\lambda_0}{n\theta^2} = k w_0^2$$



w_0 beam waist radius
n index of refraction
θ far-field diffraction angle

$\xi = \frac{2(z-f)}{b}$ normalized coordinate along z

Nonlinear Polarization

Newly generated field via processes I, II, III

$$P_4(\mathbf{r})=\frac{3}{2}N\chi^{(3)}(-\omega_4;\omega_1,\omega_2,\omega_3)E_1(\mathbf{r})E_2(\mathbf{r})E_3(\mathbf{r})$$

$$P_4(\mathbf{r})=\frac{3}{2}N\chi^{(3)}(-\omega_4;\omega_1,\omega_2,-\omega_3)E_1(\mathbf{r})E_2(\mathbf{r})E_3^*(\mathbf{r})$$

$$P_4(\mathbf{r})=\frac{3}{2}N\chi^{(3)}(-\omega_4;\omega_1,-\omega_2,-\omega_3)E_1(\mathbf{r})E_2^*(\mathbf{r})E_3^*(\mathbf{r})$$

χ the polarizability (per atom) and
N density of medium
Note degeneracy factors (field amplitudes);
those may differ for degenerate fields.

Insert fields with their mode structure:

$$P_4(\mathbf{r})=\frac{3}{2}N\chi^{(3)}(-\omega_4;\omega_1,\omega_2,\omega_3)E_{10}E_{20}E_{30}\frac{\exp(ik'z)}{(1+i\xi)^3}\exp\left[\frac{-k'\left(x^2+y^2\right)}{b\left(1+i\xi^2\right)}\right] \qquad k'=k_1+k_2+k_3=k''$$

Process I

$$P_4(\mathbf{r})=\frac{3}{2}N\chi^{(3)}(-\omega_4;\omega_1,\omega_2,-\omega_3)E_{10}E_{20}E_{30}\frac{\exp(ik'z)}{(1+i\xi)^2(1-i\xi)}\exp\left[\frac{\left(-k''+i\xi k'\right)\left(x^2+y^2\right)}{b\left(1+\xi^2\right)}\right] \qquad k'=k_1+k_2-k_3$$

Process II

$$P_4(\mathbf{r})=\frac{3}{2}N\chi^{(3)}(-\omega_4;\omega_1,-\omega_2,-\omega_3)E_{10}E_{20}E_{30}\frac{\exp(ik'z)}{(1+i\xi)(1-i\xi)^2}\exp\left[\frac{\left(-k''+i\xi k'\right)\left(x^2+y^2\right)}{b\left(1+\xi^2\right)}\right] \qquad k'=k_1-k_2-k_3$$

Process III

Further steps; Maxwell's equations

Derive amplitudes in a Fourier decomposition; amplitude with wave vector $\mathbf{K}$

$$P_4(\mathbf{K}) = (2\pi)^{-3} \int_{-\infty}^{\infty} dx'' \int_{-\infty}^{\infty} dy'' \int_{-\infty}^{\infty} dz'' P_4(\mathbf{r}'') \exp[-i\mathbf{K}\cdot\mathbf{r}'']$$

Insert in Maxwell's equation:

$$\nabla \times \nabla \times E_{4(\mathbf{K})}(\mathbf{r}) - k_4^2 E_{4(\mathbf{K})}(\mathbf{r}) = 4\pi k_0^2 P_4(\mathbf{K}) \exp[i\mathbf{K}\cdot\mathbf{r}]$$

medium vacuum

For: $\Delta k = k_4 - k'$

Solutions, process I (similar equations for II and III)

$$E_4(\mathbf{r}) = i\frac{3N}{2k_4}\pi k_0^2 b \chi^{(3)}(-\omega_4;\omega_1,\omega_2,\omega_3)E_{10}E_{20}E_{30}\frac{\exp(ik'z)}{(1+i\xi)}$$
$$\exp\left[\frac{-k'(x^2+y^2)}{b(1+i\xi)}\right]\int_{-\zeta}^{\xi}\frac{\exp[-(ib/2)\Delta k(\xi'-\xi)]}{(1+i\xi')^2}d\xi'$$

With integration boundaries:

$$\zeta = 2f/b \qquad \xi = \frac{2(z-f)}{b}$$

Field $E_4(\mathbf{r})$ to be calculated, integration over x and y, and z to z=L.

$$\int_0^{\infty} 2\pi R |E_4(R)|^2 dR \qquad R = \sqrt{x^2+y^2}$$

$\longrightarrow$

Result, generally:
- Modes cylindrically symmetric around z-axis
- Only for Process I the generated beam has lowest order Gaussian

The phase-matching can be quantified:

Phase-matching in $\chi^{(3)}$ Processes

Define the phase-matching integrals:

$$F_j\left(b\Delta k,\frac{b}{L},\frac{f}{L},\frac{k''}{k'}\right)=\frac{8}{9}\frac{k_4^2k'}{\pi^3k_0^4}\frac{1}{b^3\chi^2\left|E_{10}E_{20}E_{30}\right|^2}\int_0^\infty 2\pi R\left|E_4(R)\right|^2 dR$$

For all three processes, j=I,II,III

Dimensionless parameters, defining geometry and phases.

Then generated Power at P_4 is

$$P_4=\eta\frac{k_0^4k_1k_2k_3}{k_4^2k'}N^2\chi^2P_1P_2P_3F_j\left(b\Delta k,\frac{b}{L},\frac{f}{L},\frac{k''}{k'}\right)$$

Degeneracy factor

Phase-matching integral for Process I

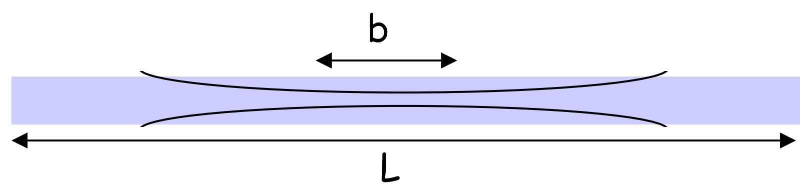
$$F_I\left(b\Delta k,\frac{b}{L},\frac{f}{L},\frac{k''}{k'}\right)=\left|\int_{-\zeta}^{\xi}\frac{\exp[-(ib/2)\Delta k\xi']}{(1+i\xi')^2}d\xi'\right|$$

This integral can be evaluated for various conditions, for example in the "tight-focusing limit", b<<L and integrating

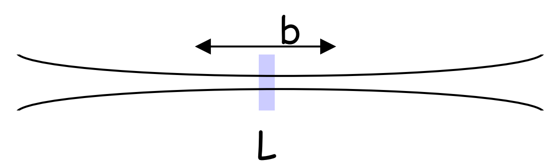
$\xi\to\infty$ $\qquad$ $\zeta\to\infty$

Define:

Tight-focus



Plane wave limit



Phase-matching integrals F_j for processes and geometries

For "tight-focus" $b/L << 1$
"half-way" the cell $f/L = 0.5$

Process I:

$$F_I(b\Delta k, 0, 0.5, 1) = \begin{cases} \pi^2 (b\Delta k)^2 e^{(b\Delta k/2)} & \text{—— } \Delta k < 0 \\ 0 & \text{—— } \Delta k \geq 0 \end{cases}$$

Similarly for Process II and III
(if $k' = k''$ assumed; lowest order mode)

$$F_{II}(b\Delta k, 0, 0.5, 1) = \pi^2 e^{(-b|\Delta k|)}$$

$$F_{III}(b\Delta k, 0, 0.5, 1) = \begin{cases} 0 & \text{—— } \Delta k < 0 \\ \pi^2 (b\Delta k)^2 e^{(b\Delta k/2)} & \text{—— } \Delta k \geq 0 \end{cases}$$

General conclusion:

THG (Process I) only possible,
in the tight-focusing limit if $\Delta k < 0$

So: only if the medium has
NEGATIVE DISPERSION !!

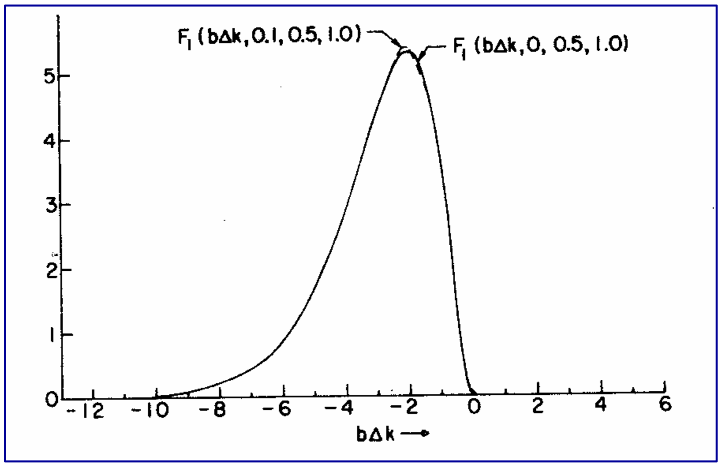
This also holds for sum-frequency mixing

Difference frequency mixing for any k

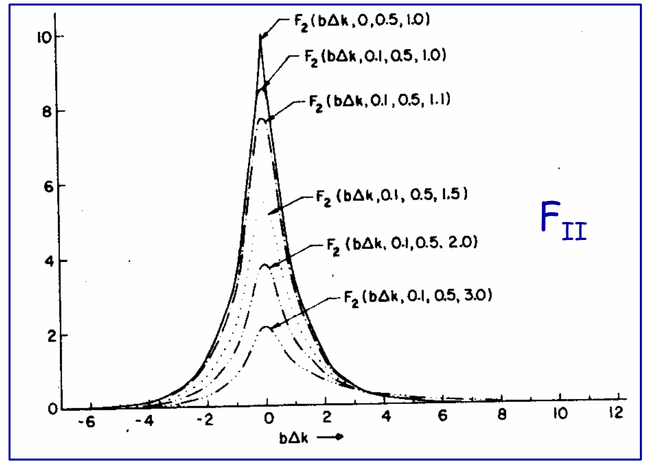
Numerical approach evaluating F_j; all near tight focus

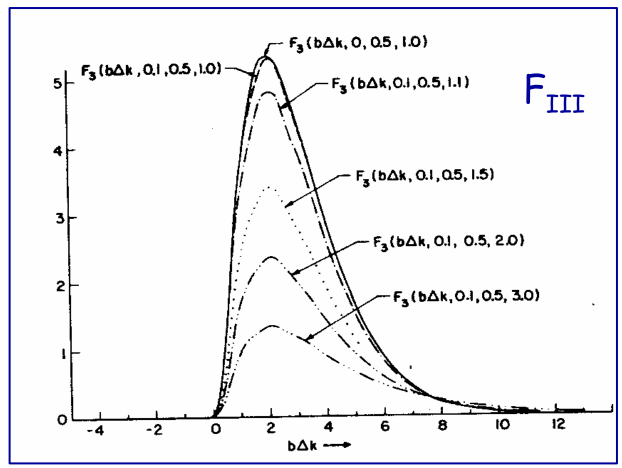
Phase-matching integral including
1) Dispersion
2) Phase evolution through focal region

$$F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)$$



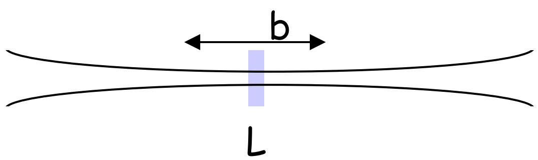
F_I versus bΔk for b/L < 0.1 and f/L=0.5



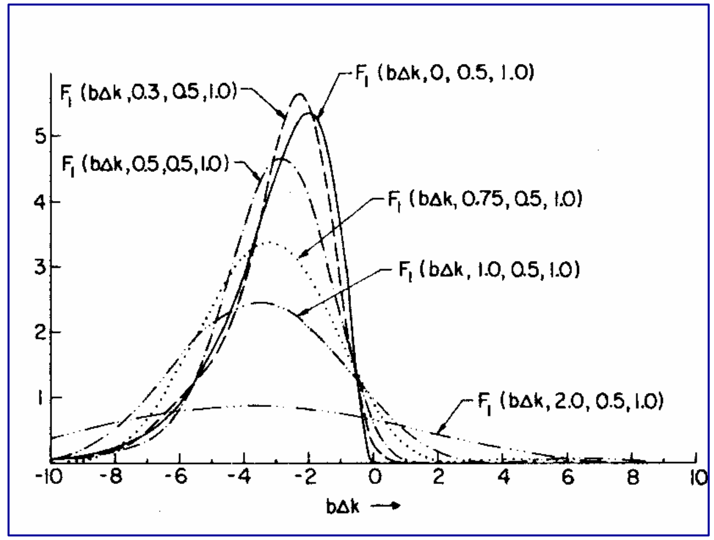


Varying the tightness of focus b/L and the mode function k″/k′

Results in the "plane-wave" limit



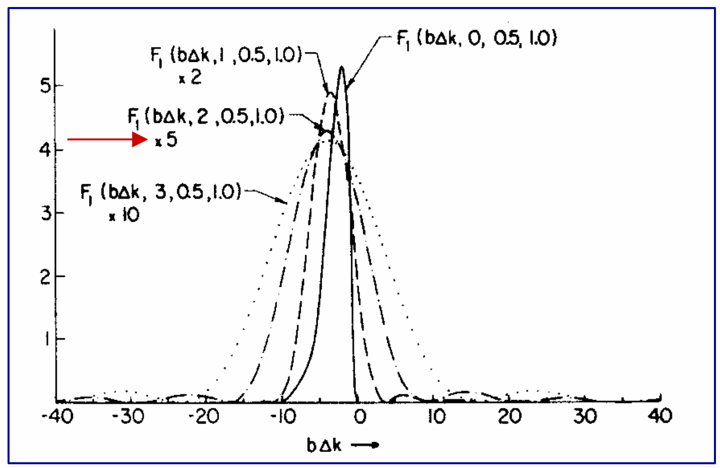
$$\lim_{b/L\to\infty} F_I\left(b\Delta k, \frac{b}{L}, 0.5, \frac{k''}{k'}\right) = \frac{4L^2}{b^2}\,\mathrm{sinc}^2\left(\frac{\Delta k L}{2}\right)$$



Note the similarity with analysis of frequency-doubling in crystal;
This was done for plane parallel fields !

At b/L ~3 the peak has shifted to the real plane-wave limit already, peaking at $\Delta k=0$.

Note also the decrease in intensity for the plane-wave limit case.



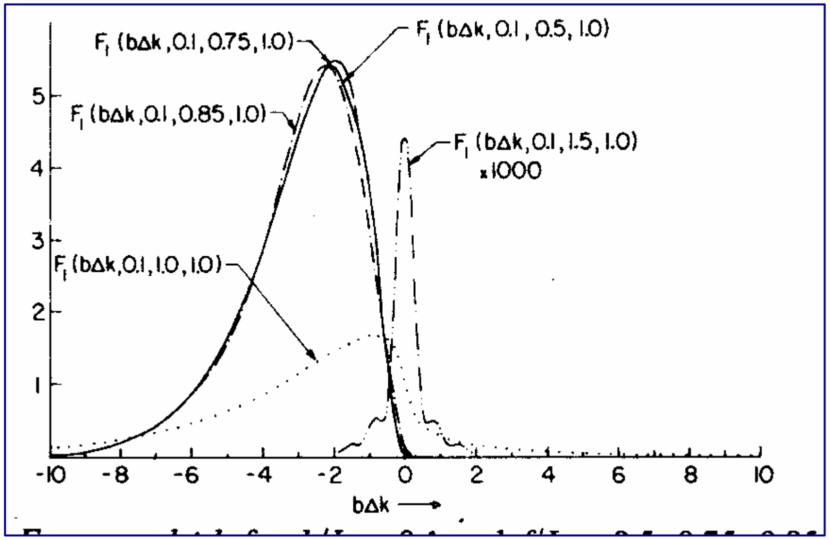
Tight-focusing is more efficient !

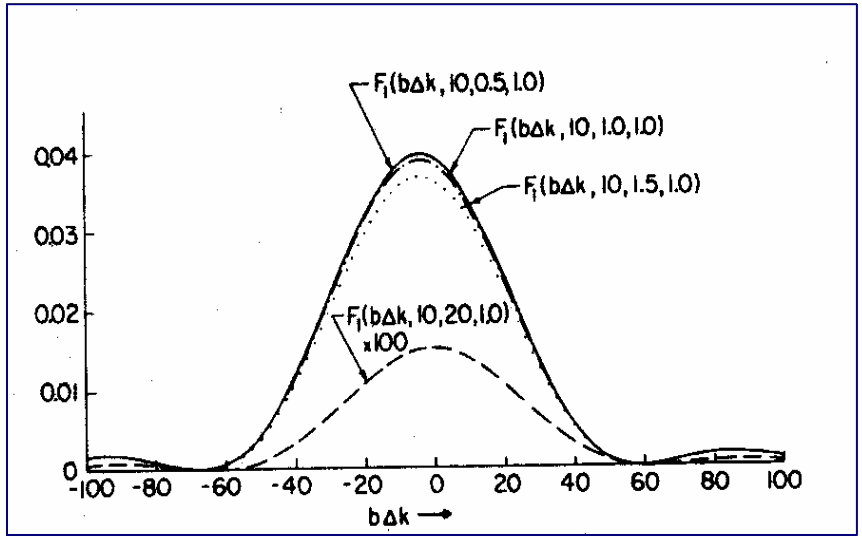
Shifting the focus; playing with the phase build-up

$$F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)$$

For near-tight-focusing of b/L=0.1
Focal positions f/L=0.5 toward f/L=1.5
In latter case focus shifts outside the cell
And effectively plane-wave limit is reached.

Near the true plane-wave limit at b/L=10
Only small changes to F-integral as long as
focus is inside the cell.
At f/L=20 the focus is twice the distance
of the confocal parameter b/L=10.





Physical interpretation of phase-matching integrals

1) Non-linear polarization peaks at the region of the beam WAIST
→ intensity is highest in focus

2) Phase-matching relates to phase-overlap of all beams involved
For a Gaussian beam:

$$\mathbf{E}_n(\mathbf{r}) = \mathbf{E}_{n0}(\mathbf{r}) \frac{\exp(ik_n z)}{1+i\xi} \exp\left[\frac{-k_n\left(x^2+y^2\right)}{b(1+i\xi)}\right]$$

$$\frac{1}{1+i\xi} = \frac{1}{\sqrt{\left(1+\xi^2\right)}} e^{-i\arctan\xi}$$

For tight-focusing:

$$\xi = -2 \longrightarrow z = f - b$$

$$\xi = 2 \longrightarrow z = f + b$$

Boundaries of focus at windows.

Lowest order Gaussian beam undergoes phase shift

$$\arctan\xi$$

when propagating through the focus (Gouy phase), adding up to π for $(-\infty, \infty)$

Driving polarization undergoes

$3\arctan\xi$	Process I
$\arctan\xi$	Process II
$-\arctan\xi$	Process III

Optimum conversion if generated field is in phase with driving fields.
Generated beam undergoes phase slip: $\arctan\xi$

In-phase for Process II, destructive for I and III

Phase-slips compensated by DISPERSION

$$\Delta k_{opt} \cong \begin{array}{cc} -2.2/b & \quad \text{I} \\ 0 & \quad \text{II} \\ 2.2/b & \quad \text{III} \end{array}$$

Optimizing density – dispersion and phase-matching

Nonlinear generation:

$$P_4 \propto P_1 P_2 P_3 N^2 \chi^2 F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)$$

$N^2 F_j$ can be optimized by varying
N, b∆k, b/L, f/L

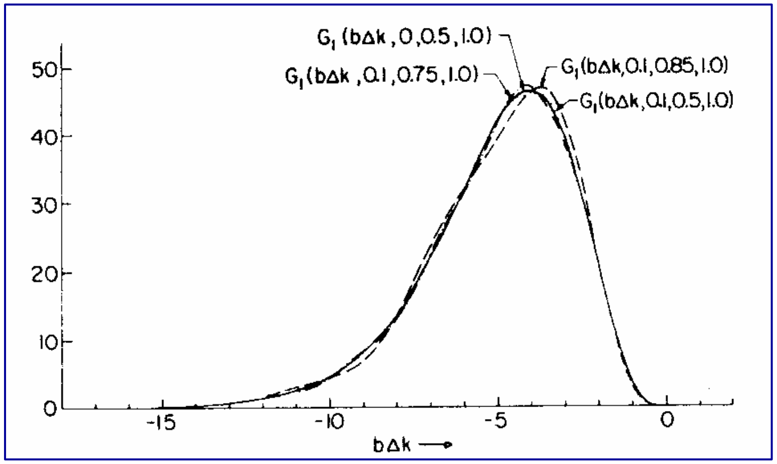
if *N* varies – macroscopic effect on medium
- index of refraction changes
- ∆k varies

Phase-mismatch is a dispersion effect and

$$\Delta k = \alpha N$$

One can define a dimensionless quantity:

$$G_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right) = (b\Delta k)^2 F_j\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)$$



G_I vs. b∆k for tight-focusing; always OK as long as focus inside the cell.

THG production and dispersion in the noble gases

Third harmonic produced:

$$\mathrm{P}_{THG} = \frac{\eta}{(3\lambda)^4} N^2 \left[\chi^{(3)}(\lambda)\right]^2 \mathrm{P}_1^3 F_j\left(b\Delta k, \frac{b}{L}, 0.5, 1\right)$$

In tight-focusing limit (b<<L):

$$F_I(b\Delta k, 0, 0.5, 1) = \begin{cases} \pi^2 (b\Delta k)^2 e^{(b\Delta k/2)} & \Delta k < 0 \\ 0 & \Delta k \geq 0 \end{cases}$$

In plane-wave limit (b>>L):

$$F_I(b\Delta k, \infty, 0.5, 1) = \frac{4L^2}{b^2} \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right)$$

We have seen : tight focus is more efficient, but then negative dispersion required

In region of bound states; index of refraction (with f_i the oscillator strength of transitions)

$$(n-1)_{lines} = \frac{Nr_e}{2\pi} \sum_i \frac{f_i}{\lambda_i^{-2} - \lambda}$$

In the continuum (σ ionization cross section)

$$(n-1)_{continuum} = \frac{N}{2\pi} \int \frac{\sigma}{\overline{\nu}_i^2 - \overline{\nu}^2} d\overline{\nu}_i$$

Cross sections and oscillator strength relate to atomic structure, with C the phase mismatch per atom;

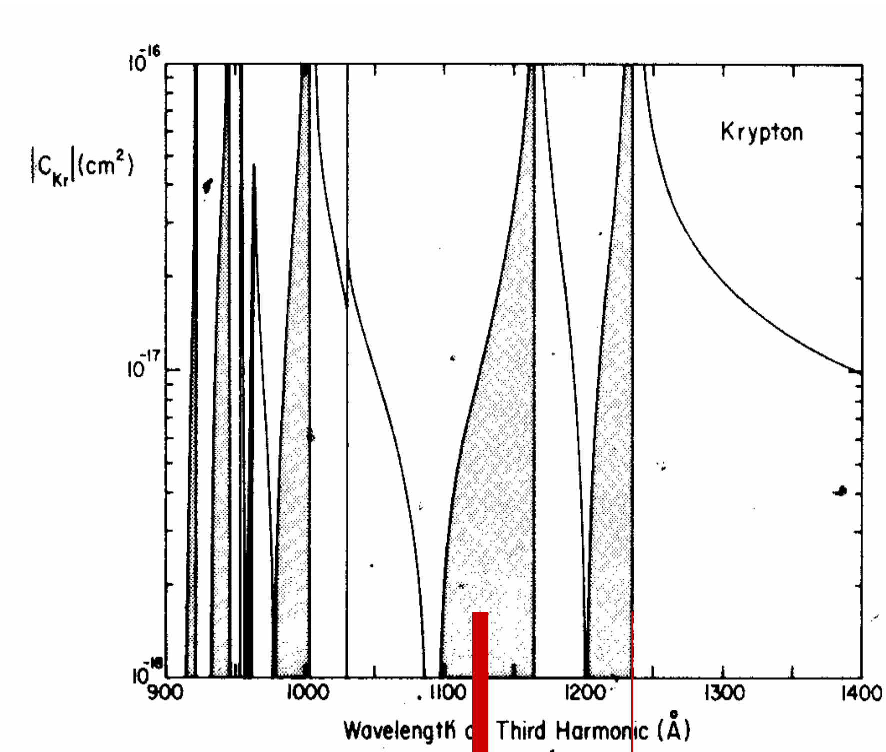
$$\Delta k = CN = \frac{2\pi(n_1 - n_3)}{\lambda}$$

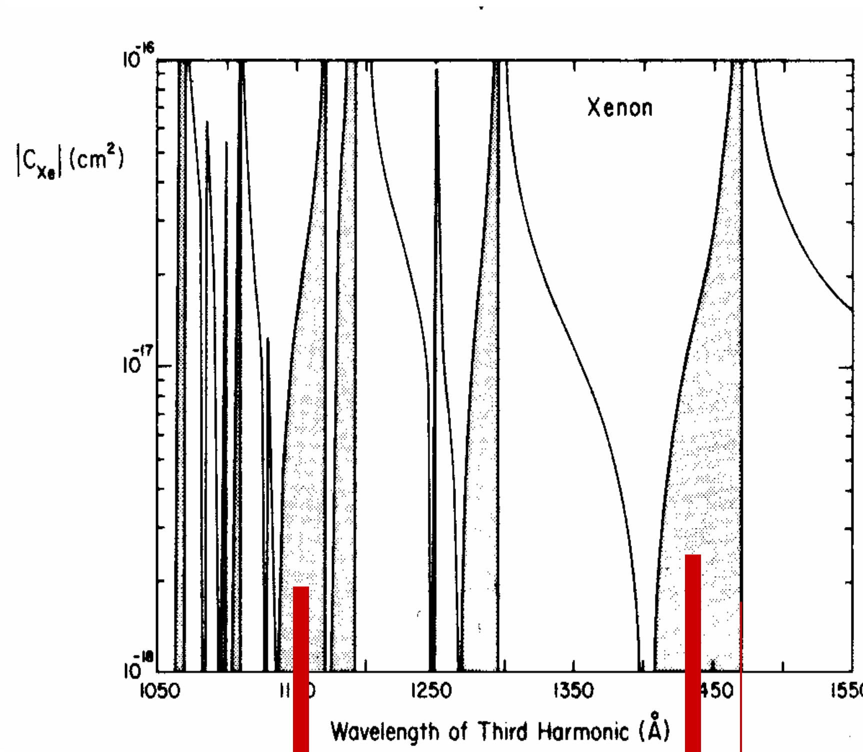
$\longrightarrow$

Calculation of regions of positive and negative dispersion

Negative and positive dispersion in the Kr and Xe

Shaded areas – "anomalous dispersion in the medium"

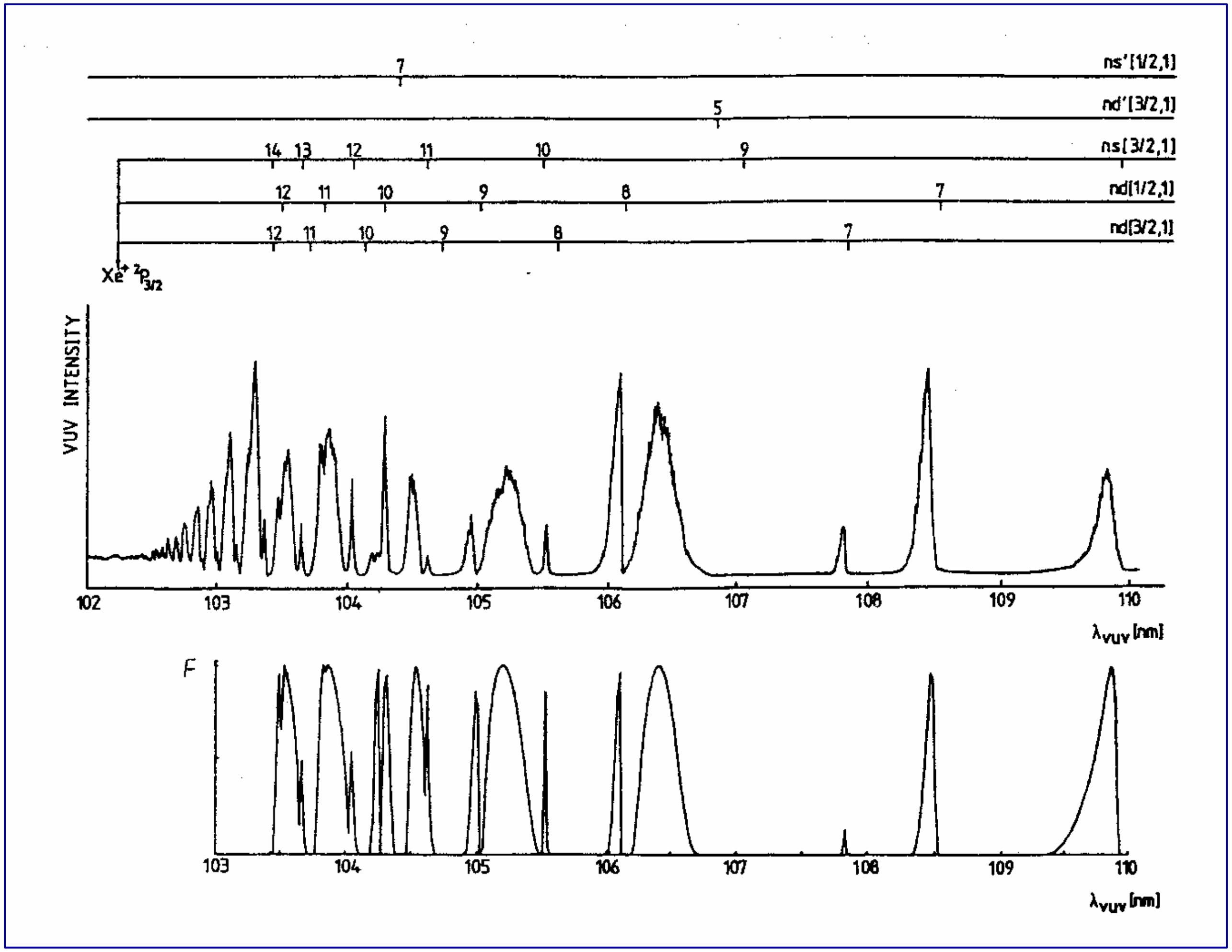




first resonance line

Regions of efficient THG

THG in Xe; experiment

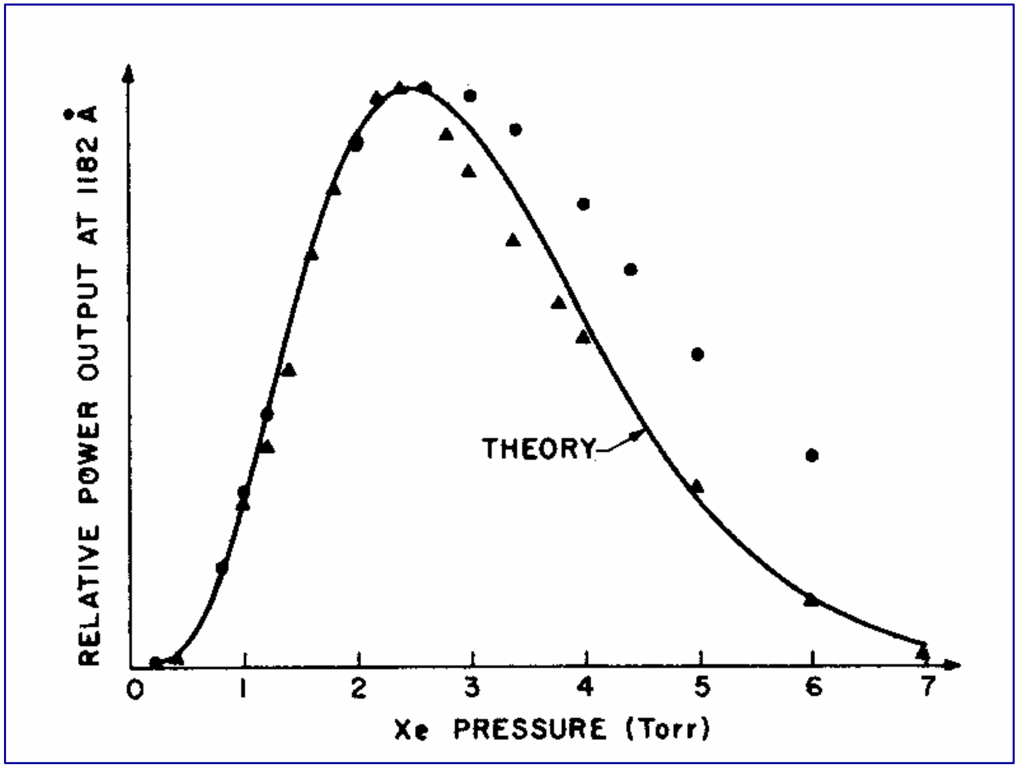


Experiment THG production

Calculation of Phase-matchingintegral

Conclusion: 1) theory of phase-matching works
2) THG effective at blue side of ns and nd resonances

Experimental density effect on phase matching in Xe

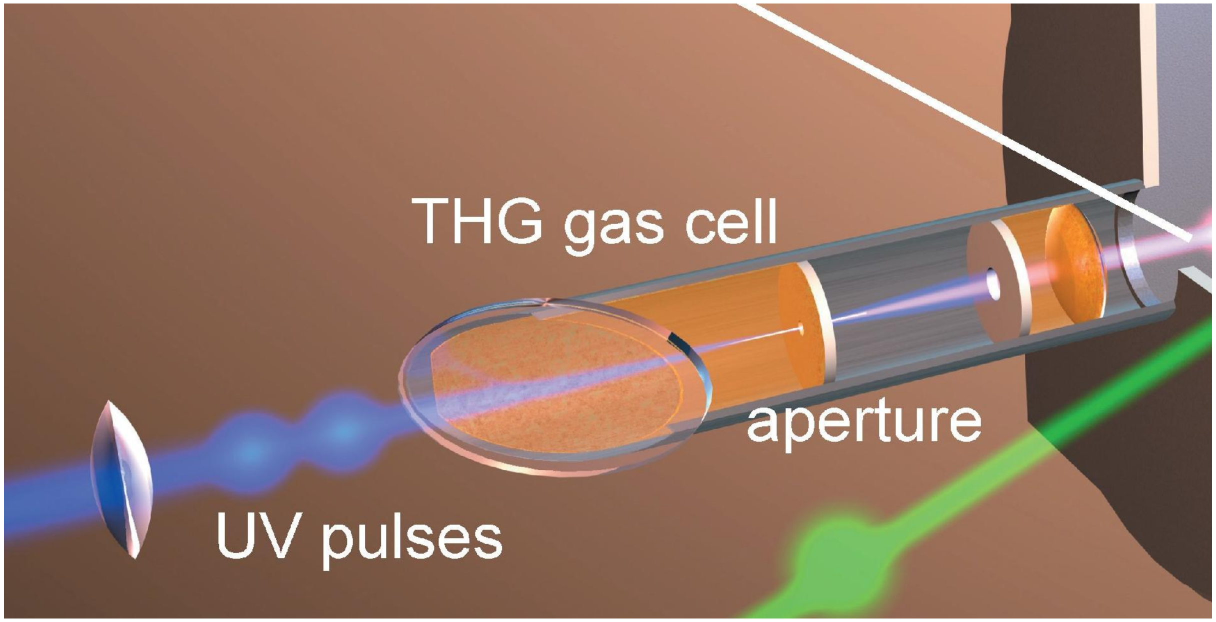


Tight focusing at λ = 118 nm in Xe
(negative dispersion);
b/L = 0.025
f/L =0.5 – centre of cell

Effect of G-integral and interplay
between N and Δk.

Defeating the negative dispersion problem

All calculations performed for integral from –L to +L.
Cut-off the medium at f; stop the destructive interference.
Can be done in "forbidden region".
Efficiency remains low; but not zero.



Resonance enhanced VUV production

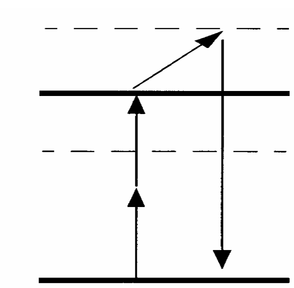
Non-linear susceptibility:

$$\chi^{(3)} \propto \sum_{gijk} \sum_{terms} \frac{\langle g|\mathbf{r}|i\rangle\langle i|\mathbf{r}|j\rangle\langle j|\mathbf{r}|k\rangle\langle k|\mathbf{r}|g\rangle}{(\omega_{gi} \pm \omega)(\omega_{gj} \pm 2\omega)(\omega_{gk} \pm 3\omega)}$$

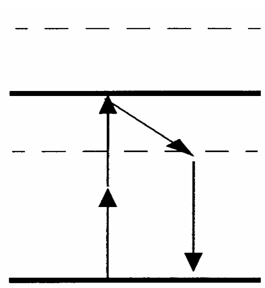
Resonances possible at the
- One photon
- Two-photon
- Three photon levels

Advantage at two-photon-level

$$\chi^{(3)} \propto \sum_{ik} \sum_{terms} \frac{\langle g|\mathbf{r}|i\rangle\langle i|\mathbf{r}|J'\rangle\langle J'|\mathbf{r}|k\rangle\langle k|\mathbf{r}|g\rangle}{(\omega_{gi} \pm \omega)(\omega_{gj} - 2\omega - i\Gamma)(\omega_{gk} \pm 3\omega)}$$



Process I



Process II

Again level structure of the noble gases is favorable:

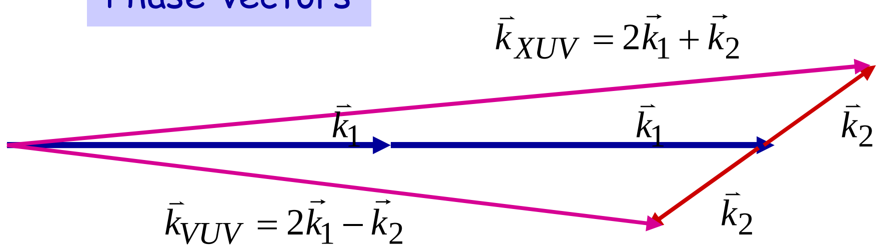
	Two-photon resonance	excitation energy (cm^{-1})	resonance wavelength (nm)	relative efficiency[a]
	5p - 6p' $[3/2]_2$	89162.9	224.3	40
	$[1/2]_0$	89860.5	222.6	187
	7p $[5/2]_2$	88352.2	226.4	4
Xe	$[3/2]_2$	88687.0	225.5	9
	$[1/2]_0$	88842.8	225.1	26
	8p $[5/2]_2$	92221.9	216.9	9
	$[3/2]_2$	92371.4	216.5	5
	$[1/2]_0$	92555.7	216.1	119
	4p - 5p $[5/2]_2$	92308.2	216.7	167
Kr	$[3/2]_2$	93124.1	214.8	56
	$[1/2]_0$	94093.7	212.6	1000

[a] Fundamental power is 20 kW.

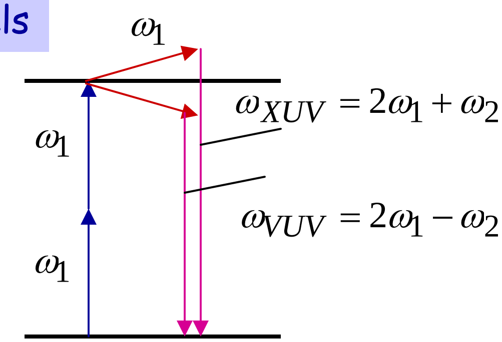
λ = 212.5 nm resonance in Kr
"strongest two-photon resonance in nature"

Non-colinear Phase-matching for sum-frequency generation

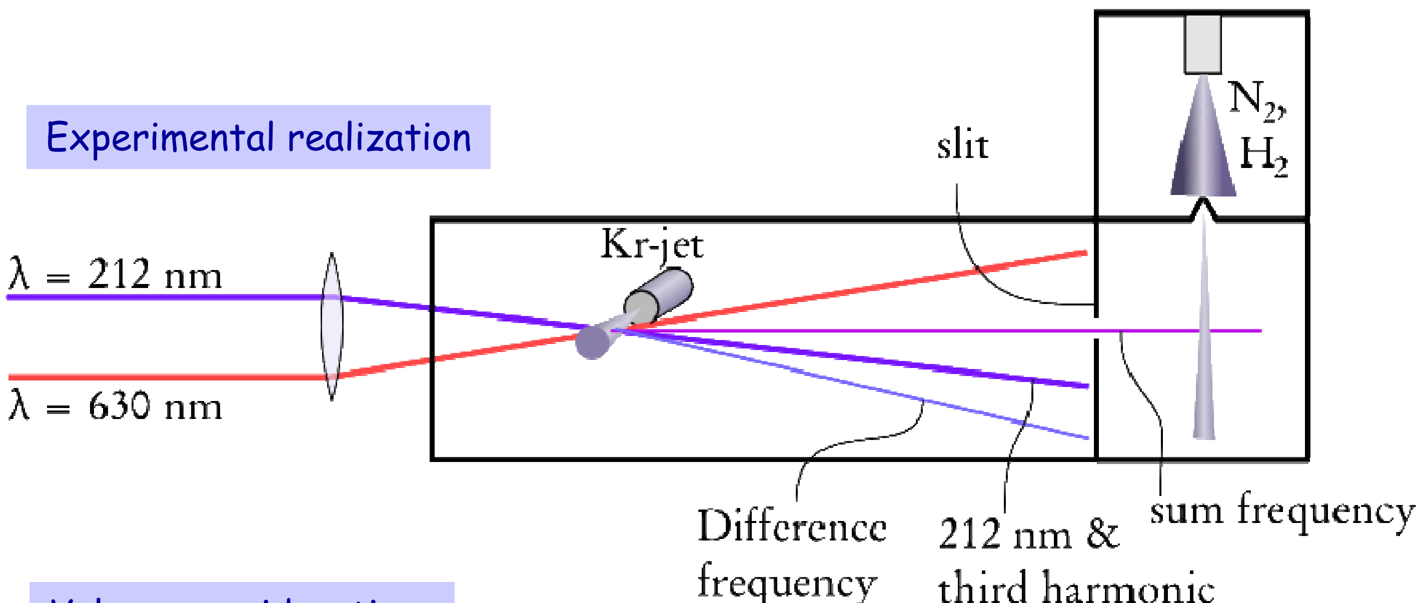
Phase-vectors



Energy levels



Experimental realization



Volume considerations

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Polarization properties

Tensor nature of susceptibility

$$\mathbf{P}_\sigma(\omega_r;\omega_i,\omega_j,\omega_k) \propto \chi^{(3)}_{\sigma\alpha\beta\gamma}\mathbf{E}^i_\alpha\mathbf{E}^j_\beta\mathbf{E}^k_\gamma$$

Dependent on matrix dipole moments

$$\chi^{(3)} \propto \sum_{gijk}\sum_{terms}\frac{\langle g|\mathbf{r}|i\rangle\langle i|\mathbf{r}|j\rangle\langle j|\mathbf{r}|k\rangle\langle k|\mathbf{r}|g\rangle}{(\omega_{gi}-\omega)(\omega_{gj}-2\omega)(\omega_{gk}-3\omega)}$$

Note: a COHERENT sum should be taken
(as in multi-photon transitions)

Parametric and non-parametric processes;
relate to energy exchange

Use atomic physics framework: evaluate
Transition dipole moments with Wigner-Eckhart
theorem:

$$\langle g|\mathbf{r}|i\rangle = \left\langle J_g M_g \left|\mathbf{r}^{(1)}_q\right| J_i M_i\right\rangle = (-)^{J_i-M_i}\begin{pmatrix} J_i & 1 & J_g \\ -M_i & q & M_g \end{pmatrix}\left\langle J_g \left\|\mathbf{r}^{(1)}\right\| J_i\right\rangle$$

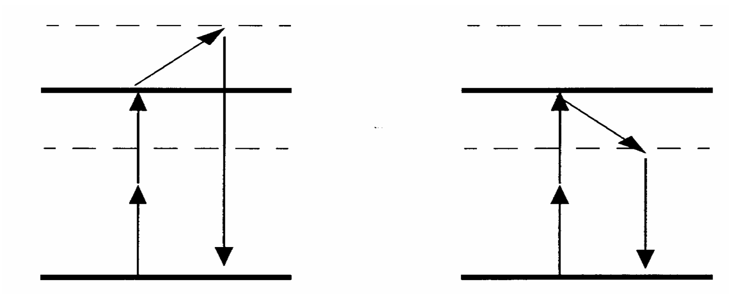
$$\chi^{(3)} \propto \begin{pmatrix} J_i & 1 & J_g \\ -M_i & q_1 & M_g \end{pmatrix}\begin{pmatrix} J_j & 1 & J_i \\ -M_j & q_2 & M_i \end{pmatrix}\begin{pmatrix} J_k & 1 & J_j \\ -M_k & q_3 & M_j \end{pmatrix}\begin{pmatrix} J_g & 1 & J_k \\ -M_g & q_4 & M_k \end{pmatrix}$$

The coherent sum requires ΔM=0 over four-photon cycle;
q=0, q=-1, q=1 projections of dipole moment on angular basis (polarizations)
Evaluate the four-product of Wigner-3j symbols

Results: 1) all polarizations linear is possible $q_1 = q_2 = q_3 = q_4 = 0$

2) THG with circular light is NOT possible: $q_1 = q_2 = q_3 = 1$

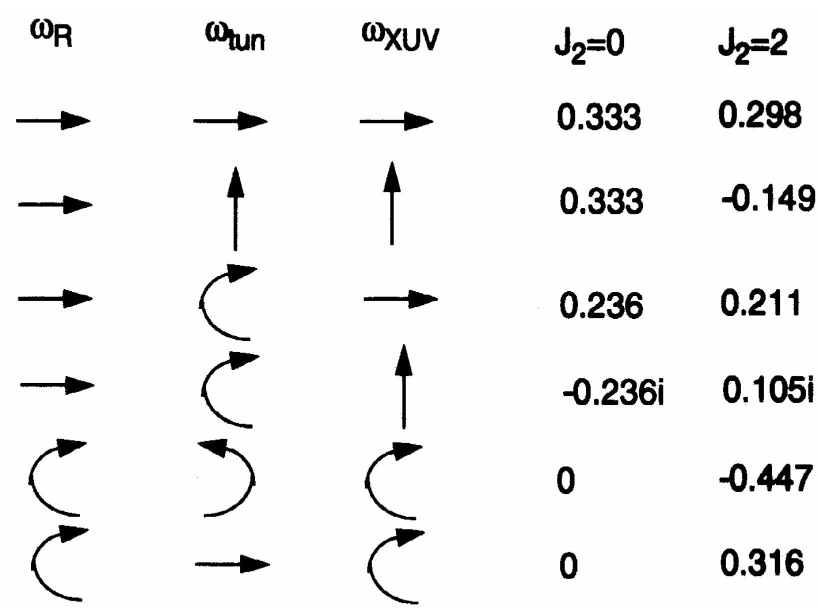
Polarization in sum-frequency mixing; quantum levels



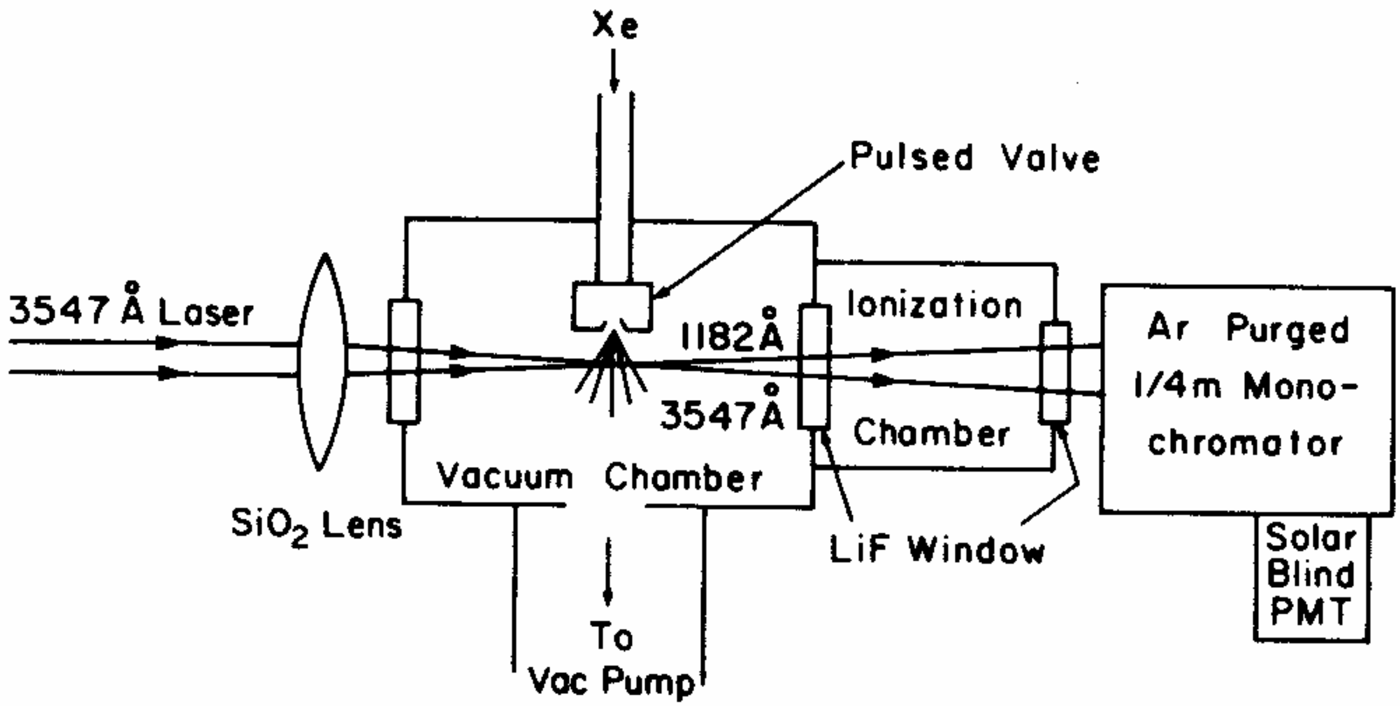
Well defined quantum level;

Real intermediate level

J_2 = 0 or 2



Pulsed jets and differential pumping – the road to the windowless regime



A.H. Kung, Opt. Lett. **8**, 24 (1983).

Some considerations

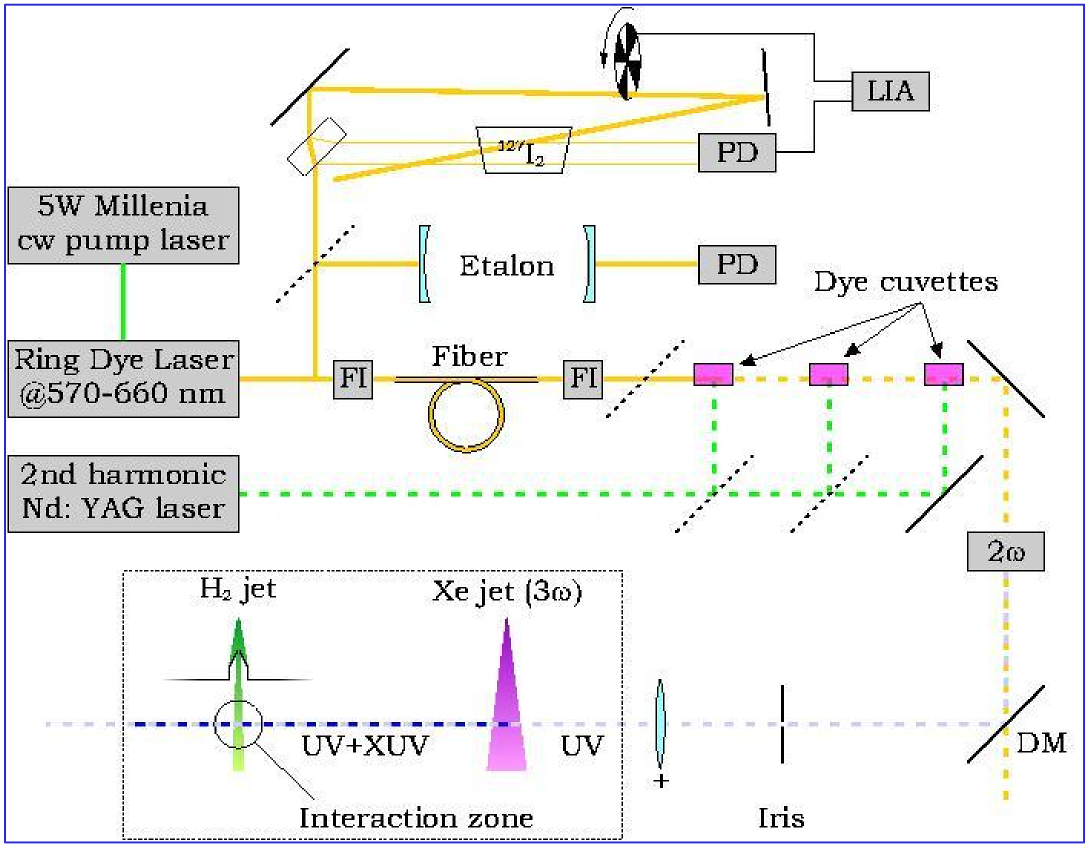
1) Bandwidth effects

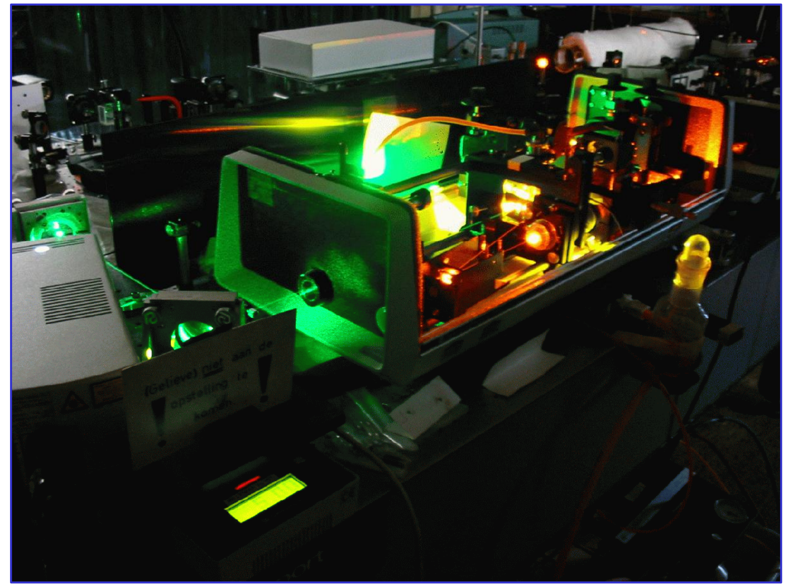
2) Lasers and synchrotrons

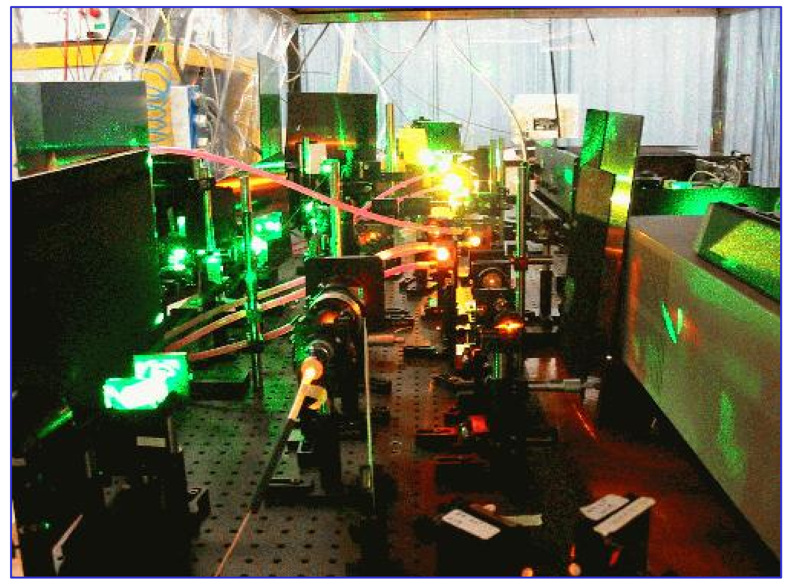
3) The perturbative regime and the non-perturbative -> recollision model

4) Spectroscopy in VUV and XUV feasible

XUV-laser setup with PDA; bandwidth ~250 MHz

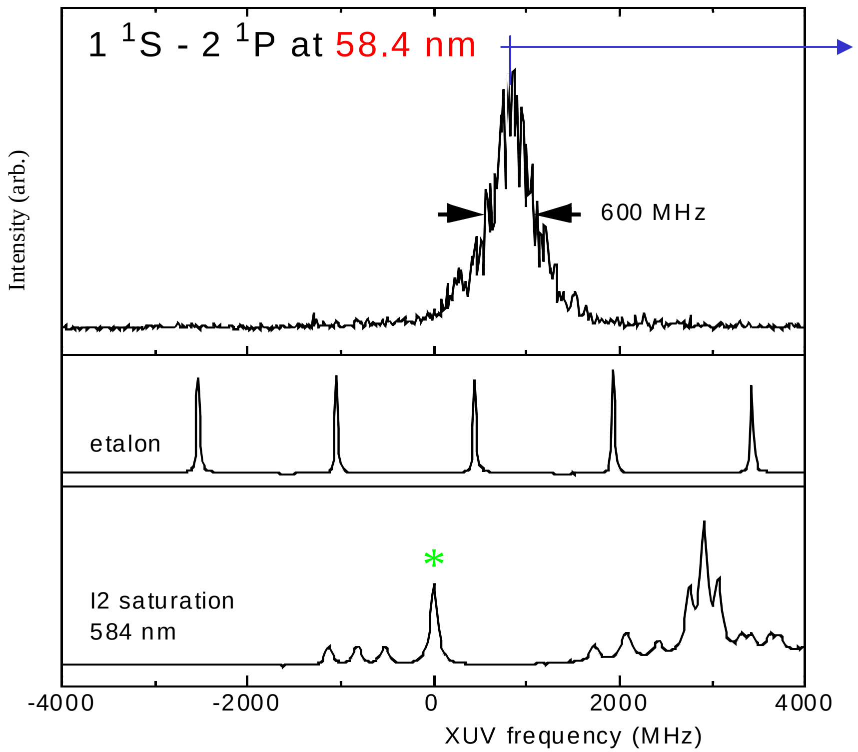






$\lambda = 90 - 110$ nm

Measurement of $1^1S – 2^1P$ resonance line of Helium



* P88(15-1) o component in I_2
at 513049427.1(1.7) MHz

5 130 495 083 (45) MHz

Results on Lamb shift in He ground state

41224 (45) MHz experiment (1997)
41233 (35-100) MHz; Drake (1993)
41223 (42) MHz theory; Korobow/Yelkovsky (2001)

2-electron QED effects
in He ($1s^2\ ^1S_0$)