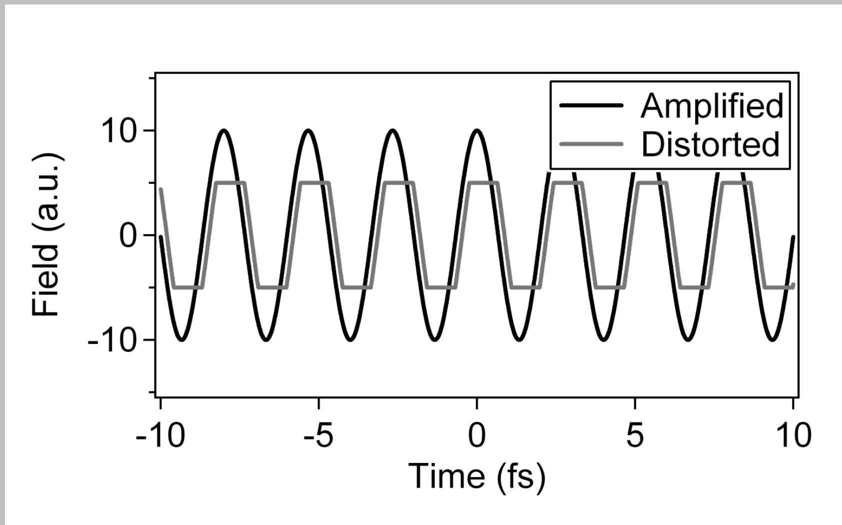
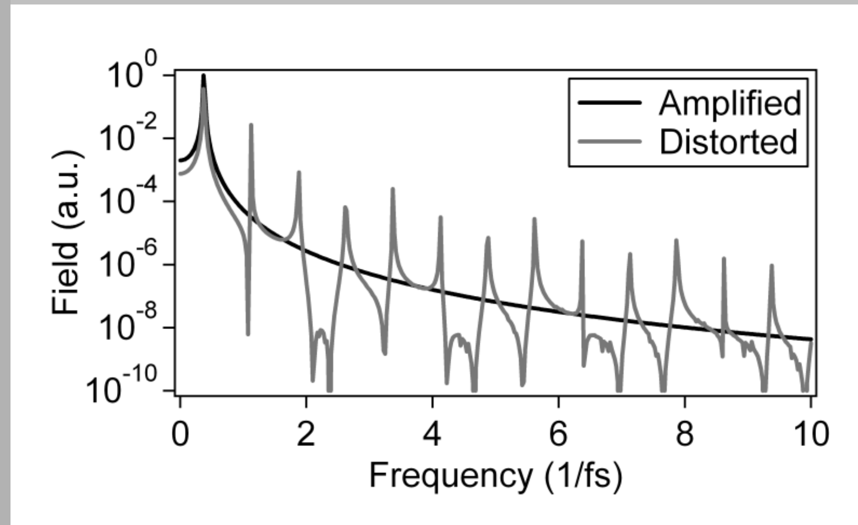
Nonlinear optics is analogous to nonlinear electronics, which we can observe easily.

Sending a high-volume sine-wave (“pure frequency”) signal into cheap speakers yields a truncated output signal, more of a square wave than a sine.
This square wave has higher frequencies: “harmonics”.



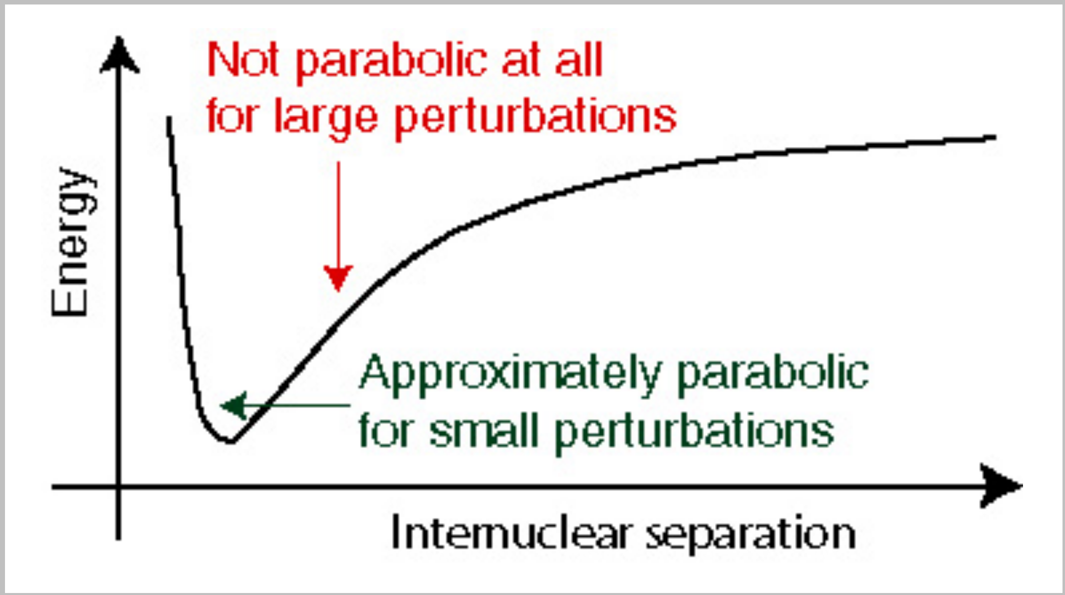


We hear this as distortion.

Nonlinear optics and anharmonic oscillators

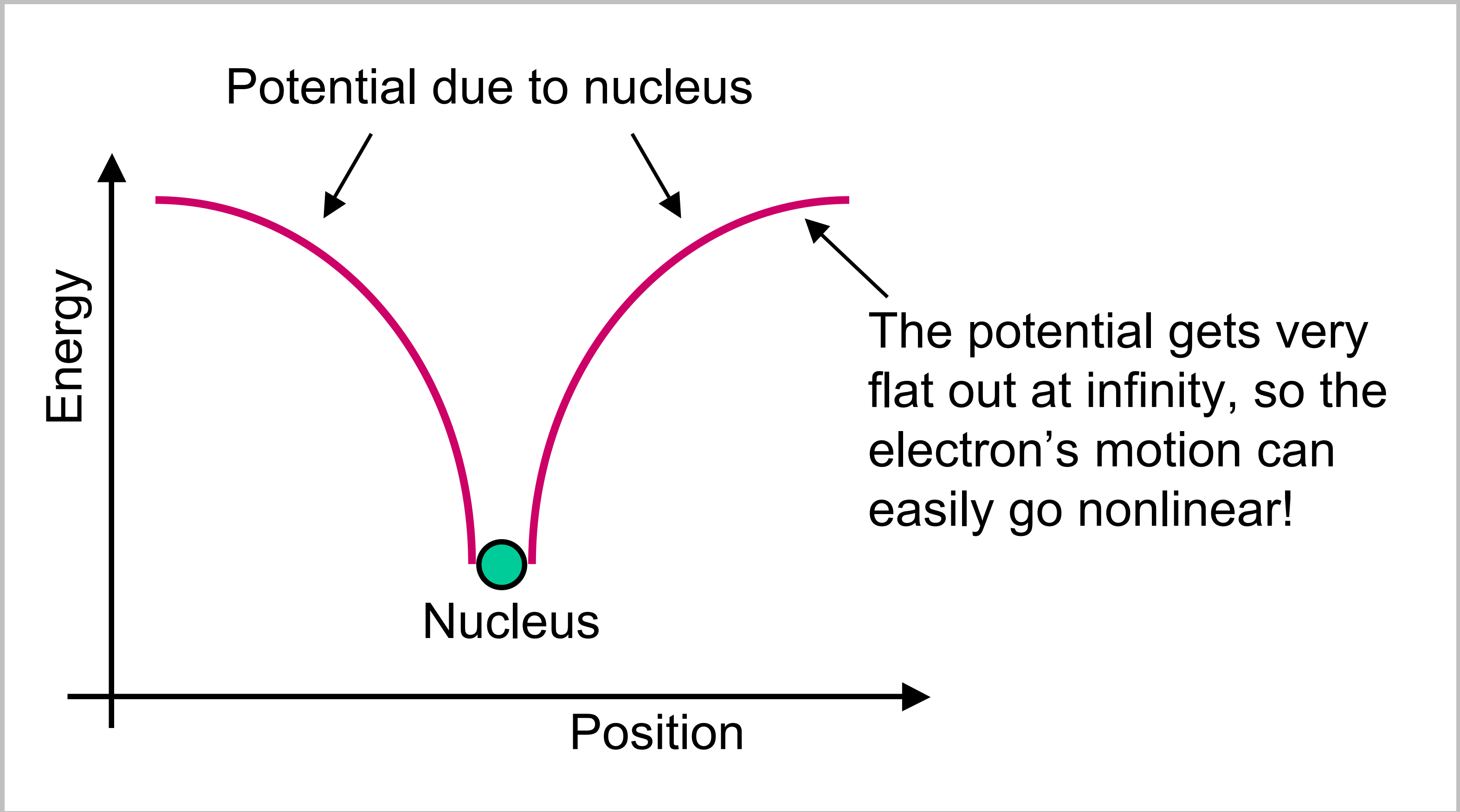
Another way to look at nonlinear optics is that the potential of the electron or nucleus (in a molecule) is not a simple harmonic potential.

Example: vibrational motion:



For weak fields, motion is harmonic, and linear optics prevails.
For strong fields (i.e., lasers), anharmonic motion occurs, and higher harmonics occur, both in the motion and the light emission.

Nonlinear effects in atoms and molecules

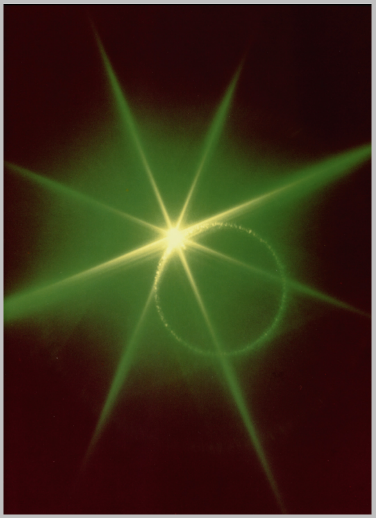


So an electron's motion will also depart from a sine wave.

Maxwell's Equations in a *Nonlinear* Medium

Nonlinear optics is what happens when the polarization is the result of higher-order (nonlinear!) terms in the field:

$$\boxed{\mathscr{P} = \varepsilon_0 \left[\chi^{(1)} \mathscr{E} + \chi^{(2)} \mathscr{E}^2 + \chi^{(3)} \mathscr{E}^3 + \ldots \right]}$$



What are the effects of such nonlinear terms?
Consider the second-order term:

Since $\mathscr{E}(t) \propto E \exp(-i\omega t) + E^* \exp(i\omega t)$,

$$\mathscr{E}(t)^2 \propto E^2 \exp(-2i\omega t) + 2|E|^2 + E^{*2} \exp(2i\omega t)$$

2ω = 2nd harmonic!

Harmonic generation is one of many exotic effects that can arise!

Sum- and difference-frequency generation

Suppose there are two different-color beams present:

$$E(t) \propto E_1 \exp(-i\omega_1 t) + E_1^* \exp(i\omega_1 t) + E_2 \exp(-i\omega_2 t) + E_2^* \exp(i\omega_2 t)$$

So:

$$E(t)^2 \propto E_1^2 \exp(-2i\omega_1 t) + E_1^{*2} \exp(2i\omega_1 t)$$ **2nd-harmonic gen**

$$+ E_2^2 \exp(-2i\omega_2 t) + E_2^{*2} \exp(2i\omega_2 t)$$ **2nd-harmonic gen**

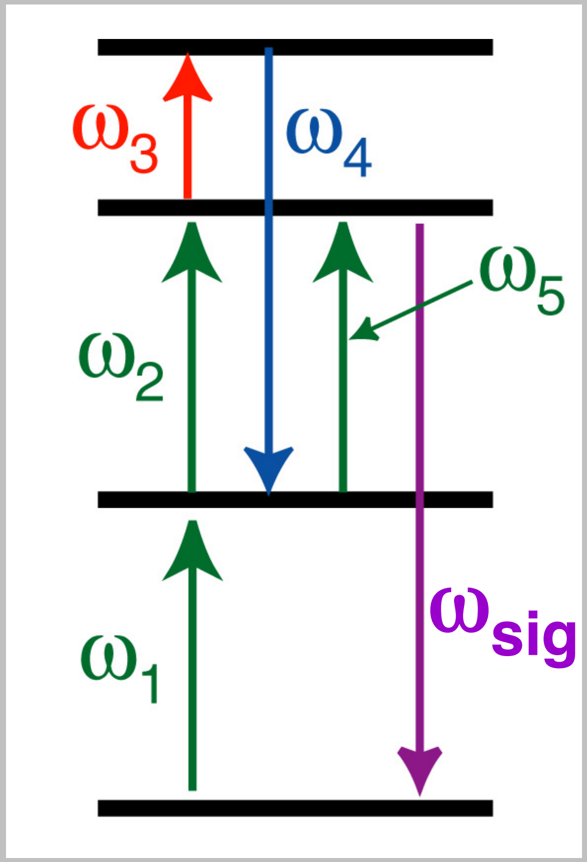
$$+ 2E_1 E_2 \exp\left[-i(\omega_1 + \omega_2)t\right] + 2E_1^* E_2^* \exp\left[i(\omega_1 + \omega_2)t\right]$$ **Sum-freq gen**

$$+ 2E_1 E_2^* \exp\left[-i(\omega_1 - \omega_2)t\right] + 2E_1^* E_2 \exp\left[i(\omega_1 - \omega_2)t\right]$$ **Diff-freq gen**

$$+ 2\left|E_1\right|^2 + 2\left|E_2\right|^2$$ **dc rectification**

Note also that, when ω_i is positive inside the exp, the E in front has a *.

Complicated nonlinear-optical effects can occur.



Emitted-light frequency

Nonlinear-optical processes are often referred to as:

"N-wave-mixing processes"

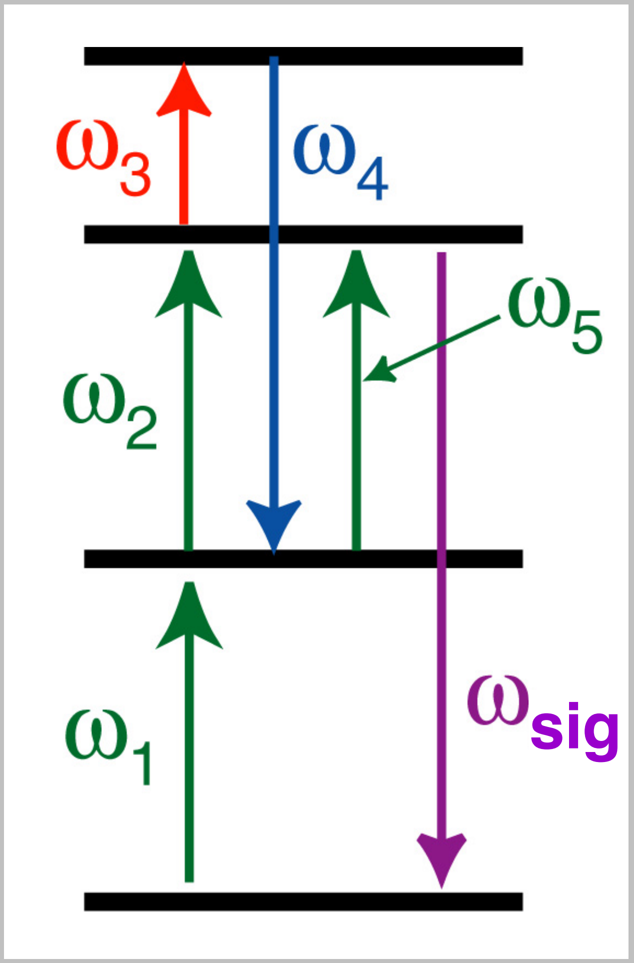
where N is the number of photons involved (including the emitted one). This is a six-wave-mixing process.

The more photons (i.e., the higher the order) the weaker the effect, however. Very-high-order effects can be seen, but they require very high irradiance. Also, if the photon energies coincide with the medium's energy levels as above, the effect will be stronger.

Induced polarization for nonlinear optical effects

Arrows pointing upward correspond to absorbed photons and contribute a factor of their field, E_i; arrows pointing downward correspond to emitted photons and contribute a factor the complex conjugate of their field:

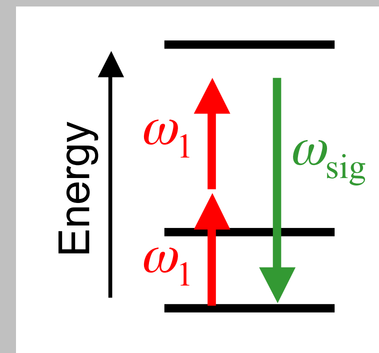
$$\mathscr{P} = \varepsilon_0 \chi^{(5)} E_1 E_2 E_3 E_4^* E_5$$



Conservation laws for photons in SHG

Energy must be conserved:

$$\omega_1 + \omega_1 = \omega_{sig} \quad \Rightarrow \quad \omega_{sig} = 2\omega_1$$

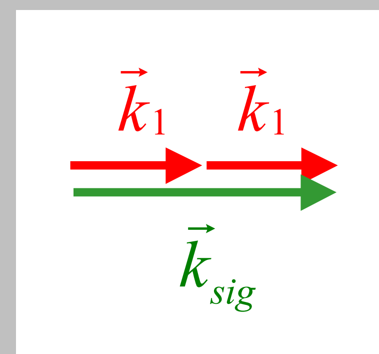


Momentum must also be conserved:

$$\vec{k}_1 + \vec{k}_1 = \vec{k}_{sig}$$

$$\omega_{sig} = 2\omega_1$$

$$\Rightarrow \quad \cancel{2\frac{\omega_1}{c_0}}\, n(\omega_1) = \cancel{\frac{2\omega_1}{c_0}}\, n(2\omega_1)$$



To simultaneously conserve energy and momentum:

$$\Rightarrow \boxed{n(\omega_1) = n(2\omega_1)}$$

The phase-matching condition for SHG!

First Demonstration of Second-Harmonic Generation

P.A. Franken, et al, Physical Review Letters 7, p. 118 (1961)

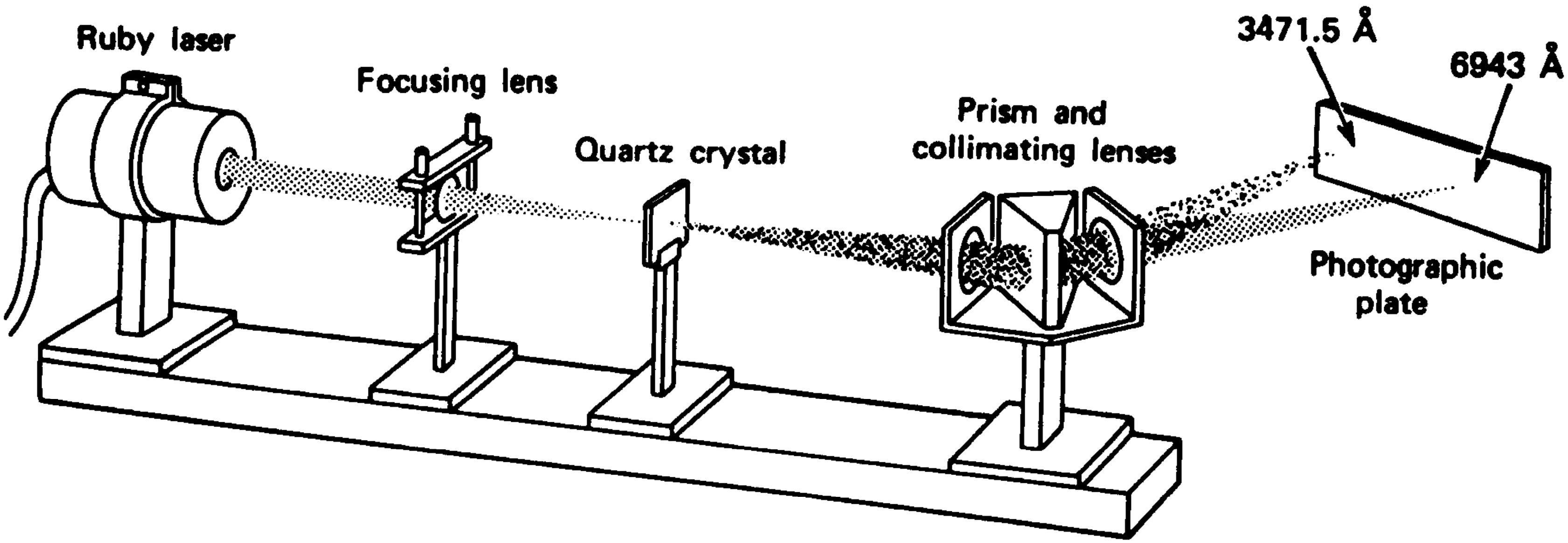


Figure 12.1. Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694\ \mu m$ is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347\ \mu m$. The two beams are then separated by a prism and detected on a photographic plate.

The second-harmonic beam was very weak because the process wasn't phase-matched.

First demonstration of SHG: The Data

The actual published result…

The second harmonic

Input beam

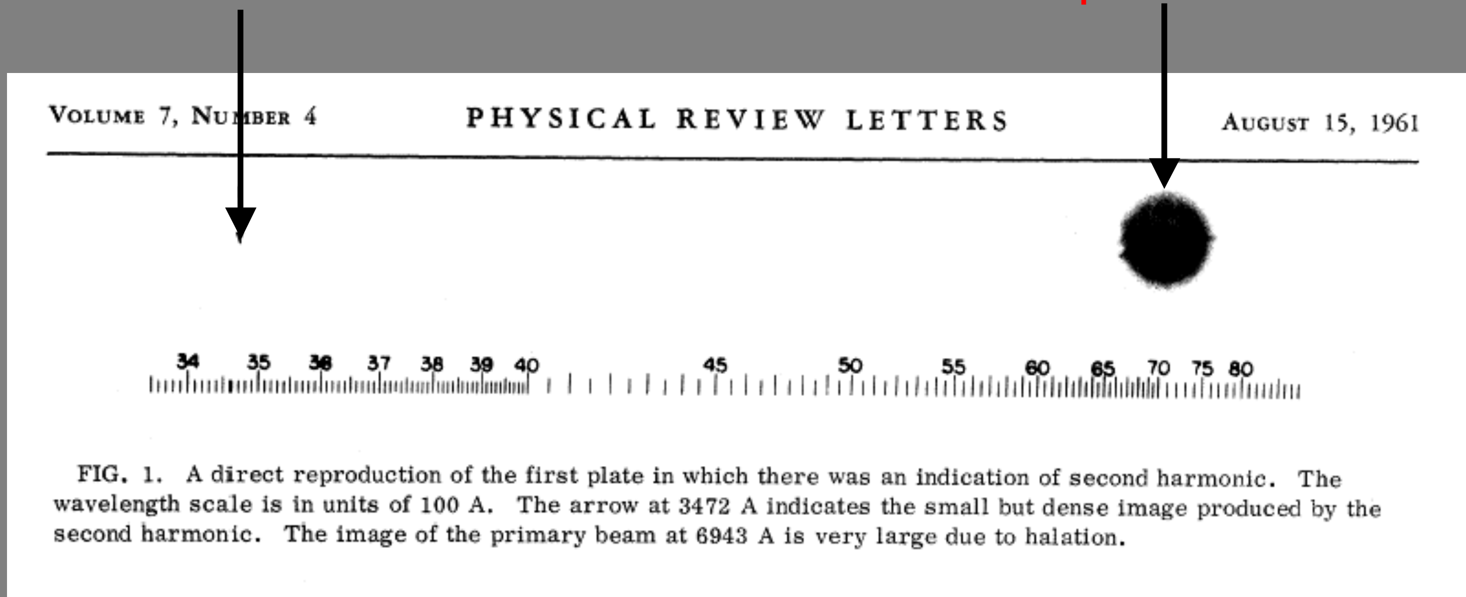
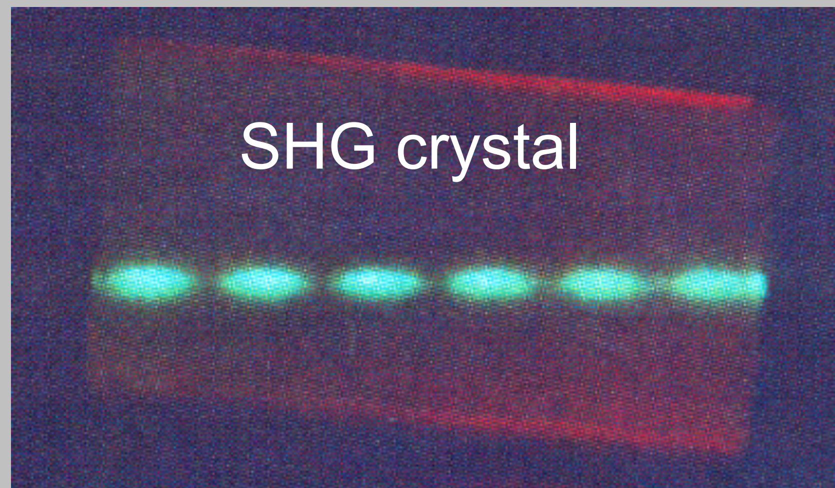


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

Note that the very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt.

Light created in real crystals

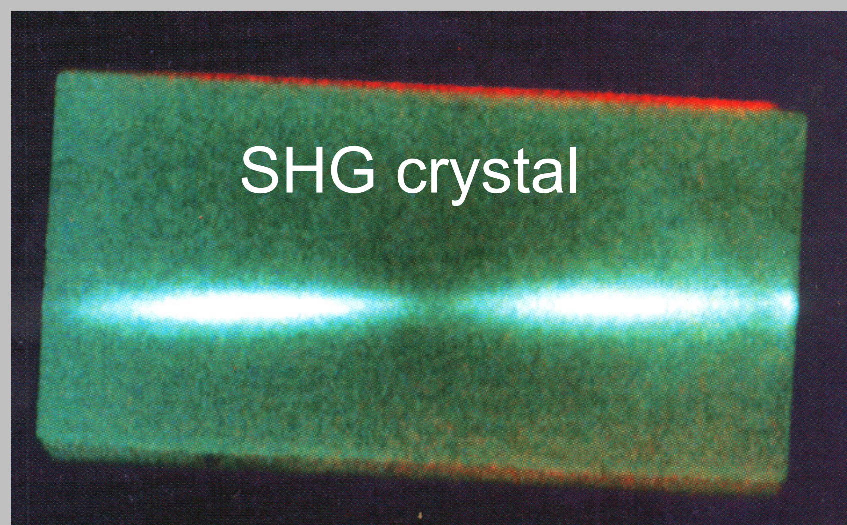
Far from
phase-matching:



Input beam

Output beam

Closer to
phase-matching:



Input beam

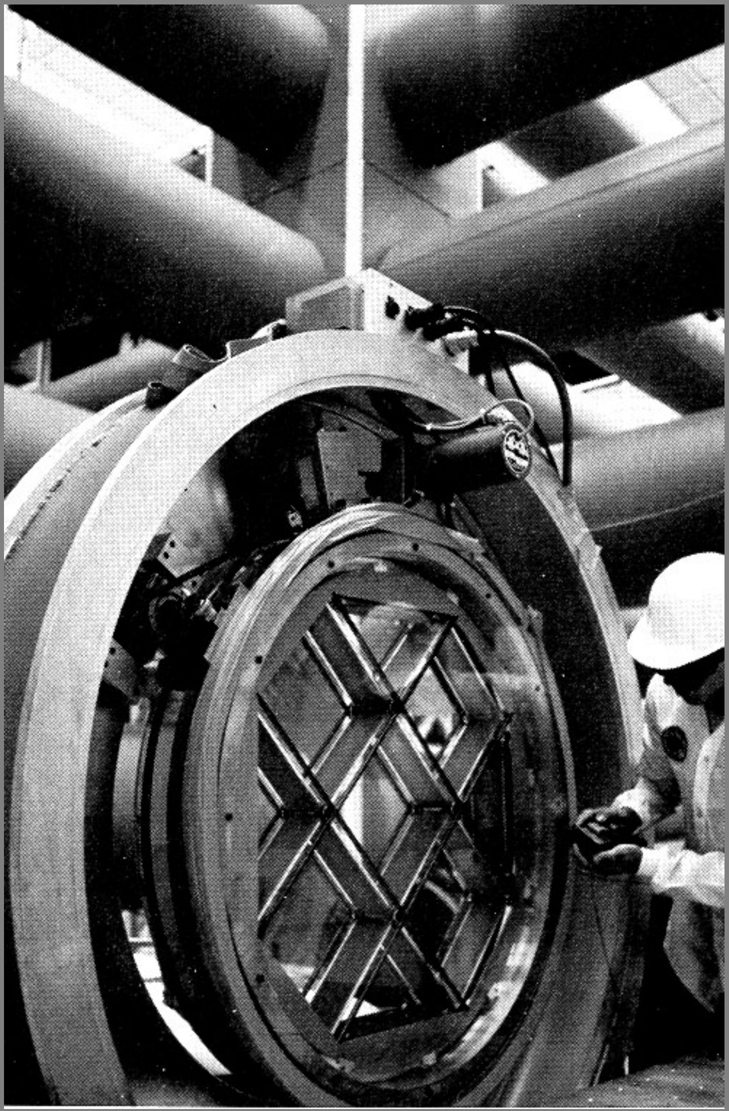
Output beam

Note that SH beam is brighter as phase-matching is achieved.

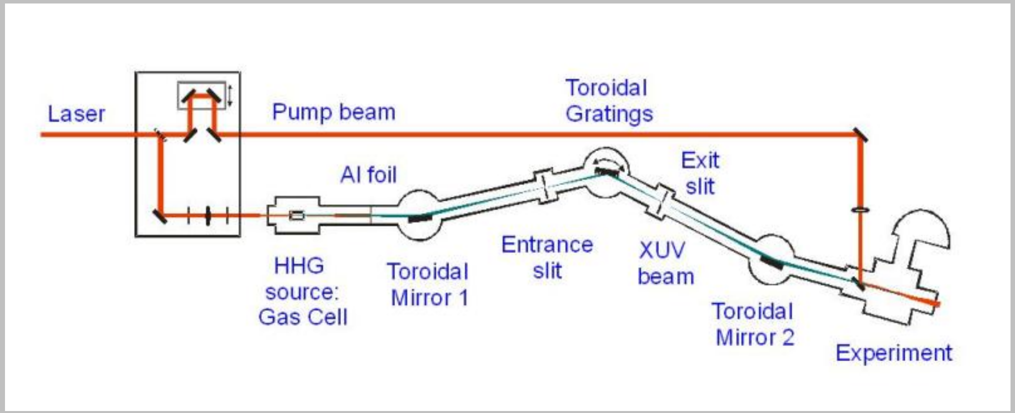
Second-Harmonic Generation

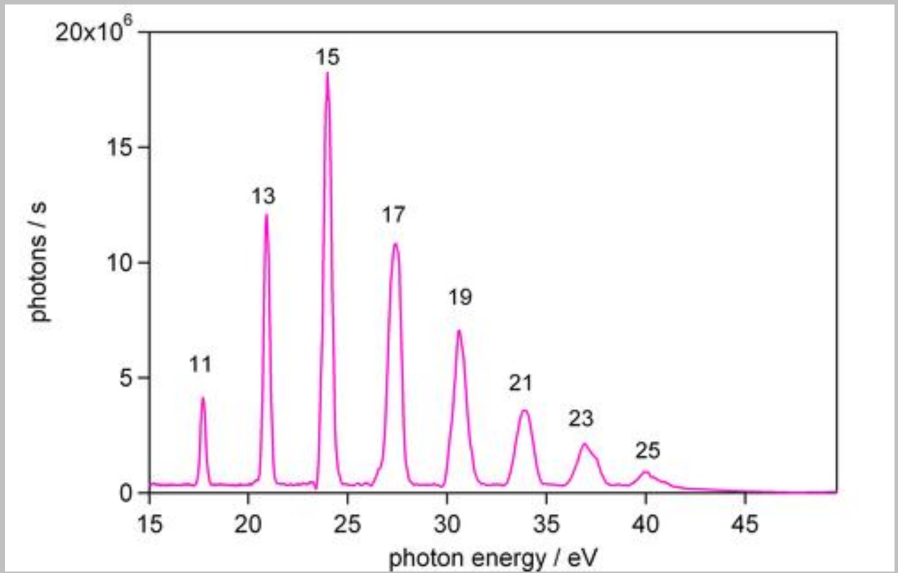
SHG KDP crystals at Lawrence Livermore National Laboratory

These crystals convert as much as 80% of the input light to its second harmonic. Then additional crystals produce the third harmonic with similar efficiency!



High-order harmonic generation





- Atoms driven with field strength comparable to binding field
- Odd harmonics only
- Current observations beyond 300th harmonic!

Images:
http://www.mbi-berlin.de/de/research/projects/4-1/subprojects/UP3/