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Efficient method for the extraction of second harmonic light after extracavity frequency doubling

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A method is given for the isolation of second harmonic radiation after extracavity frequency doubling of light from a synchronously pumped cw dye laser. The efficiency is close to 100%, and the original beam direction is restored.

In our laboratory, samples of biological interest are investigated using time-resolved fluorescence and fluorescence anisotropy.¹ The detection technique is time-correlated single photon counting, requiring high repetition rate (up to 1 MHz) picosecond light pulses with nanojoule to picojoule energies for excitation.² For this purpose a mode-locked Arion laser together with a synchronously pumped dye laser is used.

Tunable UV pulses are obtained with a temperature tuned ADA crystal,³ generating picojoule pulses at 280–310 nm wavelength from the output of a synchronously pumped rhodamine 6G dye laser. At these power levels the efficiency

of second harmonic conversion is of the order of 10^{-2} – 10^{-3} . Formerly, radiation at the fundamental wavelength was suppressed using filter glass of 2 × 2 mm thickness (Schott UG11). Transmission for the second harmonic was 50–60% for the fundamental $<10^{-7}$.

The use of a filter glass for harmonic extraction is simple and effective; however, there are some drawbacks. First, the waste of 50% of excitation power may be a problem in experimental situations. In addition, the fundamental wavelength light should not be completely absorbed by glass but may be used for the generation of a reference timing signal for the detection chain via a photodiode. Furthermore, the surface of colored glass band filters in this wavelength region will often show surface imperfections, giving rise to scattering and beam deformation. Apart from that, these absorbing filters can only safely be used in the laser beam because the average power is only a few milliwatts due to a decrease in the repetition rate of excitation pulses down to 596 kHz by means of an electrooptic modulator setup.⁵ Another problem will arise when tunable UV is generated in the 320-350 nm wavelength region, for example, by frequency doubling with LiIO₃ of the radiation of a synchronously pumped DCM dye laser. In particular, in the 340–350 nm part of that region all relevant types of filter glass become more or less transmissive for the fundamental frequency light.⁴

In principle, the orthogonality of the polarization directions of fundamental frequency and second harmonic light enables the separation of the different polarized states with a Glann laser polarizer. However, we found the fundamental frequency light after the frequency doubler to be unsufficiently linearly polarized ($<10^4/1$) to obtain satisfactory rejection for applications where a large dynamic range is required.⁶

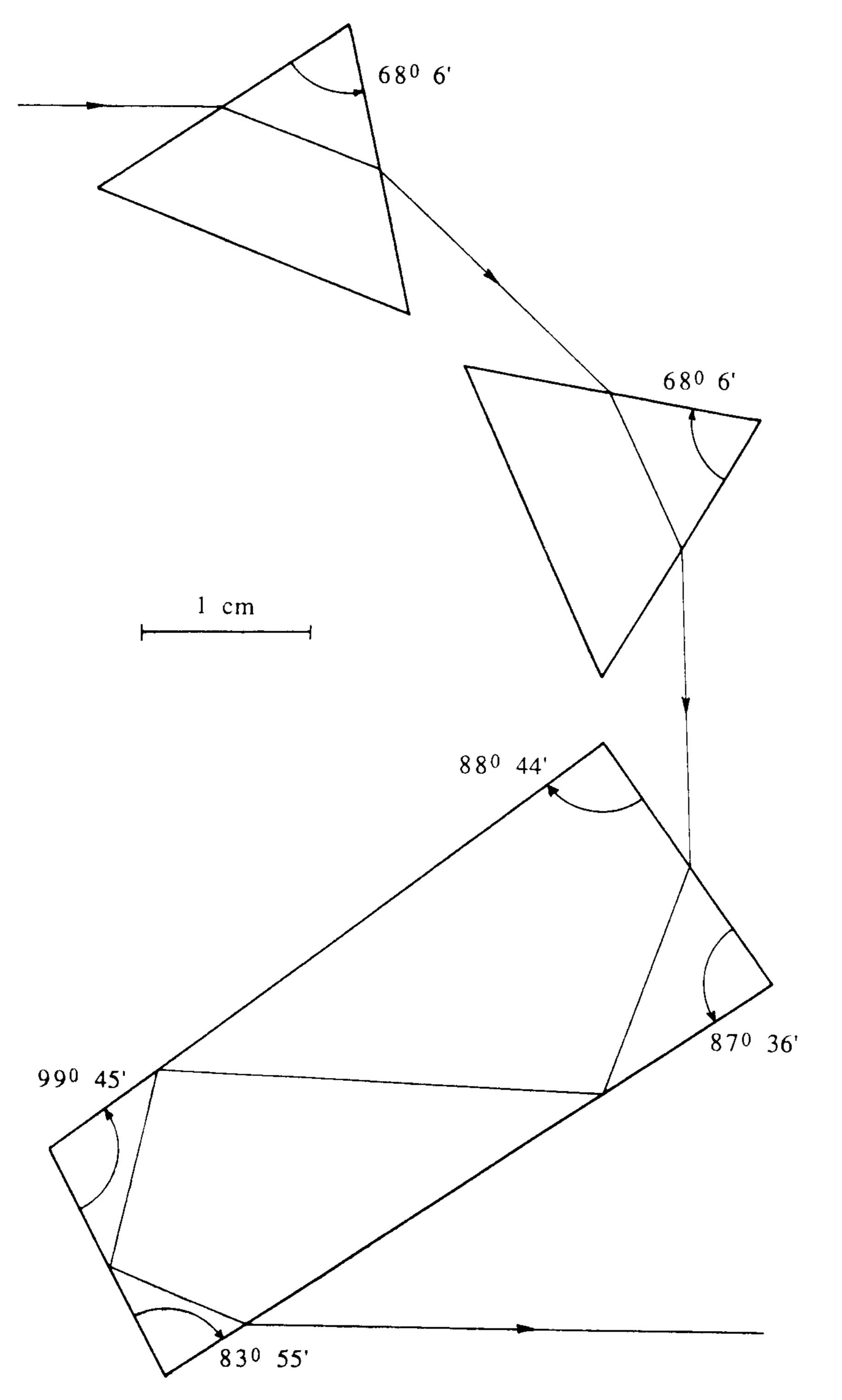


Fig. 1. Cross section of the prisms. The given pathway is that of the UV beam.

Brewster angle (reflection-loss free) refraction of polarized light⁷ is frequently applied in (electro-)optics. It is often utilized for intracavity applications to take optimum advantage of the optical gain inside laser cavities. Even a completely mirror free cavity for a cw laser has been built.⁸ Also extracavity applications are numerous. JK-lasers applies an extracavity gull-wing harmonics separator in Q-switched Nd:YAG lasers, but in this case there is abundant power available, and larger deviations from the Brewster angle are then less important.

Here a combination of prisms is described to separate first and second harmonic light after extracavity frequency doubling utilizing Brewster angle refractions. The aims for design were: a complete (>10⁷/1) suppression of fundamental frequency light, the availability of part of the fundamental frequency light for reference timing purposes, minimum losses, restoration of the directions of both the beam and polarization, matching of the difference in height between laser beam, and sample housing entrance using a minimum number of (preferably uncoated rugged easy-to-clean) components, and last but not least the design should be compact for stability and to avoid occupying a big part of the optical table surface.

The frequency doubling setup consists of two simple thin lenses (fused silica) for focusing and recollimation of the beam with the doubling crystal placed between (a temperature-tuned ADA crystal from Quantum Technology or an angle-tuned LiIO₃ crystal from Gsänger). The incoming red laser beam is horizontally polarized by the final polarizer of the pulse-picker setup.⁵

In Fig. 1 a cross section of the harmonic extraction setup is presented. The material used for these components is UV grade fused silica (Suprasil I). After harmonic generation, the more or less recollimated beam containing first and second harmonic light enters the upper prism at the Brewster angle for the vertically polarized UV light. Here a reflection

originates from the horizontally polarized first harmonic radiation. From the Fresnel formulas⁹ a reflection of 18% can be calculated. Starting with nanojoule pulses this reflection delivers $\sim 10^9$ photons per pulse. This is sufficient to generate a reference timing signal via a fast photodiode.

The first five boundary surfaces are all at the Brewster angle for the second harmonic light, giving a decrease to 35% of first harmonic power because of the orthogonality of the polarization directions of first and second harmonic. The first two prisms are used to disperse the UV and red parts of the beam. The third prism is used to restore the beam direction. At the sixth to eighth surface a total internal reflection is used; the ninth boundary surface is again at the Brewster angle for the second harmonic.

Finally, a dispersion angle of 7° between the two beams of the harmonics is created, sufficient to separate the harmonics on lateral displacement with a diaphragm within a few centimeters of the light path. The vertical displacement of the UV beam can be adjusted by moving the third prism, but there is a minimum displacement of \sim 6 cm. Although the components are designed for 335 nm, the setup can be used over a wide wavelength range. At 480 and 280 nm the reflection losses are $<4 \times 10^{-5}$ per boundary surface.

The prisms are mounted by gently screwing them via a metal lever on an adjustable vertical metal plate that was covered with a thin layer of black felt to enhance the grip on the plate for the prisms. Surrounding the optical components a thin metal box is mounted with holes just for input, output, and reference beams. Also, this box is covered inside with a thin layer of black felt to absorb red reflection beams as much as possible.

During the mounting procedure the fundamental laser beam is suppressed by a colored glass filter (Schott UG1 or UG11). The optimum angle for the Brewster angle refraction at the prisms is adjusted by observing the blue fluorescence on a piece of white paper of the reflection from the first boundary surface and rotating the prism for minimum reflection losses. For wavelengths different from the design wavelength of the prisms, the prisms are adjusted by observing the position of the main UV beam after refraction through the prism and tuning the rotation of the prisms for a minimum deflection angle. In this case refractions are equally divided between both surfaces and the losses are minimal.

The transfer efficiency of the setup is determined using a Photodyne model 66XLA optical power and energy meter with a model 400 broadband silicon photodiode sensor head. No diffèrence between input and output UV power can be measured more than the $\pm 1\%$ reproducibility of the meter system.

The first harmonic suppression is ideal, i.e., limited by scattered light radiation intensity from the silica component surfaces. This intensity is $<10^{-7}$ of the second harmonic power at a distance of 50 cm.

The first harmonic reflection from the first boundary surface of the first prism is used to generate a timing reference signal via a lens (f = 50 cm) and a HP 5082-4203 PIN photodiode (45 V reverse bias), driving the 50 Ω input of an Ortec model 934 quad constant fraction discriminator. A neutral density filter is used to prevent overload of the photodiode.

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