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Use of copper lasers for high resolution spectroscopy: generation of 10mW narrowband tunable laser light at around 210 nm

G.A. NAYLOR, I.G. KOPRINKOV, G. SITJA, H. RING and J.P. PIQUE

Laboratoire de Spectrométrie Physique, UA 008, Université Joseph Fourier de Grenoble, BP. 87, 38402 Saint-Martin-d'Hères cedex, France

Copper lasers have for a long time been used in spectroscopy to generate tunable narrowband light in the range 580 - 750 nm by pumping a dye laser and 750 - 1000 nm by pumping a titanium sapphire laser. The generation of high UV powers (~ 500 mW) in the range 290 - 375 nm has been made possible by frequency doubling in BBO, the output of a dye laser. Recently wavelengths down to 210 nm at average powers up to 10 mW have been produced by frequency mixing the doubled and fundamental output of the copper laser pumped dye laser. Such tunable light sources are finding numerous applications in linear spectroscopy due to their high average output powers but relatively low peak powers. Large signals can thus be obtained without provoking multiphotonic effects. We discuss some of these applications.

Introduction

Copper vapour lasers provide one of the most efficient ways of generating coherent light in the visible. This type of laser is characterised by a high average output power (10-100W) on two wavelengths (510.6nm and 578.2nm) in the green and yellow with a medium peak power (50-300kW) and high pulse repetition rates (5-30kHz). The visible output in the green and yellow matches well the absorption band of various tuneable laser sources used in spectroscopy, notably dye lasers (Rhodamine, DCM etc...) and titanium sapphire lasers. The visible rather than UV pump wavelengths of excimer lasers, allows much greater dye lifetimes to be achieved. Commercial copper vapour lasers are typically about 1% efficient in converting AC mains power to optical power, which compares favourably with the 0.02% efficiency typical of CW ion lasers. Recently a newly developed type of copper laser, named the copper hybrid laser [1] offers higher efficiencies still (typically 2-3%). The medium peak power and high repetition rates of the resultant output of these tuneable lasers are ideal for harmonic generation (SHG, THG) of shorter wavelengths. Such tuneable UV sources have proved to be useful for linear spectroscopy where non-linear effects in the sample are to be avoided (saturation, multi-photon ionisation, Stark broadening and shifts etc...) [1]. Recently [2] the third harmonic of a dye laser operating around 640nm has been generated by frequency doubling and mixing the dye laser to generate laser light around 213nm (see fig 1). The typical performance is shown in table 1. The advantage of such a system compared to other methods of generating coherent deep UV are:

- i) The relatively low peak power avoids non-linear effects.
- ii) The high relatively high average power gives a large signal.
- iii) The narrow band emission is comparable with the Doppler width of small molecules at ambient temperature (few GHz).

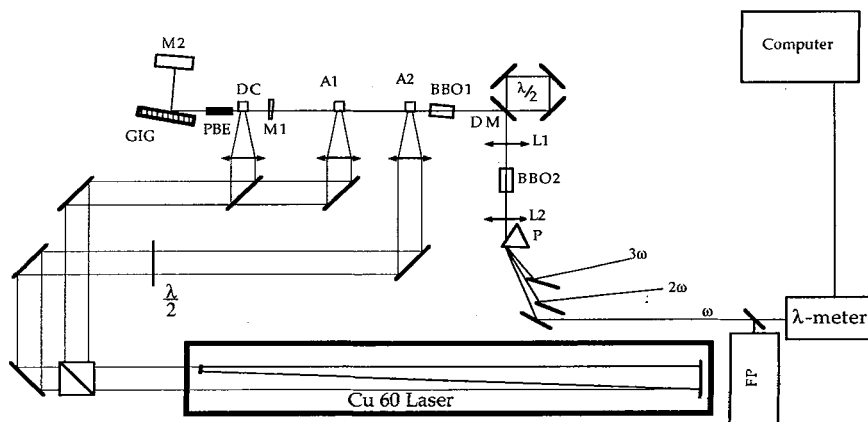


Fig. 1 : Copper laser pumped dye laser : frequency doubling and mixing.

Optical arrangement for the generation of the third harmonic

The copper laser and dye laser are shown in fig 1. The copper laser is an Oxford lasers Cu60 giving around 40-50W on both lines when fitted with an unstable cavity. The dye laser is 'home made' and consists of a Littman type oscillator [6] using a two prism beam expander with single pass dye amplifiers. Each stage uses flowing dye cells with vertically flowing dye and are all pumped with vertically polarized beams. Tuning is achieved by kinematic rotation of the plane mirror at the rear of the oscillator cavity. An average output power of 5W can be achieved with this arrangement at 640nm using DCM dye. The bandwidth is typically 2-3GHz. The DCM is mixed with a little Rhodamine 590 to increase the efficiency in the amplifiers, by increasing the absorption of the yellow pump wavelength. The output of the dye laser can be frequency doubled simply by placing a crystal of BBO just after the second amplifier. The dye laser beam at the output of the second amplifier is about 1mm in diameter and well collimated. The beam is well below the damage threshold of BBO at this point and conversion efficiency of 10% of the beam power to UV is typically achieved. Higher efficiencies should be achievable by working closer to the damage threshold using focusing optics, but our arrangement has the advantage of its relative ease of alignment and reliability. This tuneable UV around 320nm has been used in LIDFS experiments to study the quantum chaos in the vibrational modes of CS₂ [7]. A further frequency conversion step has been implemented to generate the third harmonic. Frequency mixing of the fundamental and doubled output of the dye laser is performed in a second BBO crystal cut for type 1 phase matching. The crystal used for this second step was actually cut for type 1 frequency doubling of 440nm, so had to be used at a fairly high angle of incidence, incurring high Fresnel reflection losses. For type 1 phase matching, the polarisations of the two beams to be mixed must be parallel, whereas the frequency doubled beam from the first crystal is generated with a polarisation orthogonal to the fundamental beam polarisation. The UV and red beams from the first crystal are therefore split by a dichroic beam splitter and the red (fundamental) beam is rotated 90° in polarisation by a small horizontal loop in which the beam makes a small vertical step. The two beams are then recombined using the same beamsplitter and focussed into the second BBO crystal with a 5cm focal length singlet silica lens. The output beams from the crystal were recollimated with a second similar lens and separated by a silica prism. In this way up to 10 mW at around 213nm was generated. This source provide one of the highest powers achieved for a tuneable source around this wavelength. The advantages compared to other systems using Nd:YAG or Excimer lasers are its narrow bandwidth (few GHz) coupled with its relatively low peak power (around 200W) to avoid non-linear, eg multi-photon effects. The high repetition rate (6.5kHz) ensures a high average power. This source has been used to study the pre-dissociation of CS₂ by exciting the

1B_2 state (and subsequently the generated CS from the $\chi^1\Sigma^+$ state) [2] see figs 2 and 3. Shorter wavelengths still are to be expected by using dyes that lase at shorter wavelengths (eg. the rhodamines).

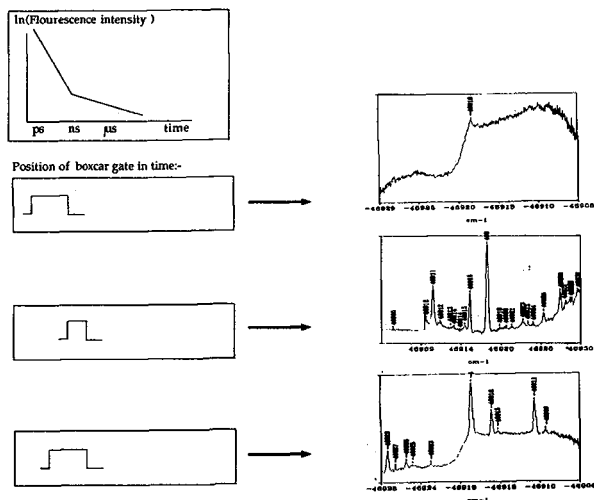


Fig. 2 : Experimental arrangement to measure total fluorescence around 213 nm.

Fig. 3 : Total fluorescence of CS_2 at different delay times following excitation pulse.

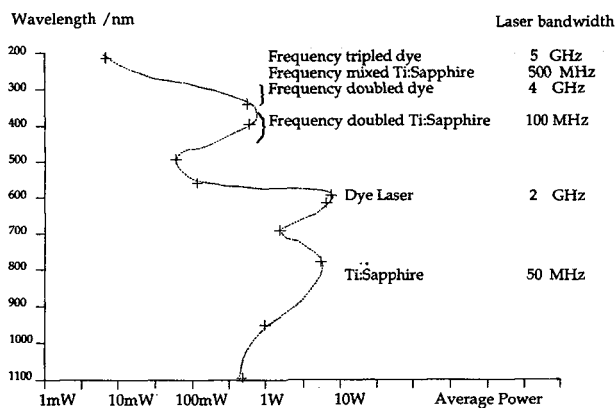
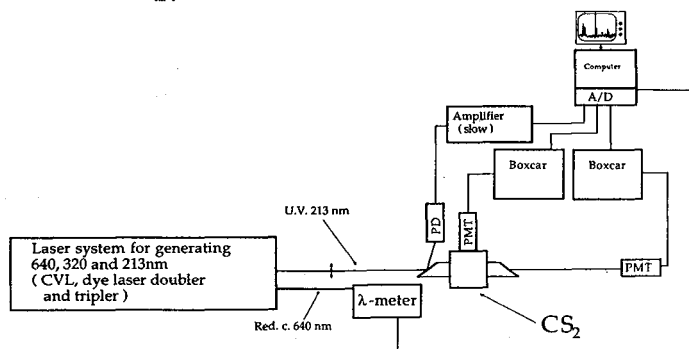


Fig. 4 : Spectral region accessible using CVL, dye laser, titanium sapphire, frequency doubling and mixing.

Pumping of titanium doped Sapphire lasers

The emission wavelengths of the copper vapour laser also match the absorption band of titanium doped sapphire. A copper laser pumped titanium sapphire laser in a Z-cavity has been used for intra-cavity stimulated emission and intra-cavity absorption [8,9]. With an intra-cavity cell containing gas excited by a second pump beam, the observation of stimulated emission pumping was also accompanied by mode-locking of the titanium sapphire laser. The output wavelength range of titanium sapphire lasers (700-1000nm) matches well available single mode diode lasers. Injection seeding of copper pumped titanium sapphire lasers offer interesting opportunities for high resolution spectroscopy (laser bandwidth below 100MHz) [10]. The output of a titanium doped sapphire laser can also be frequency doubled and mixed with the direct output of the copper laser to achieve tuneable blue and UV wavelengths.

Conclusions

Copper vapour laser pumping of dye lasers and titanium sapphire lasers coupled with frequency doubling and mixing allows a very wide wavelength range to be covered (see fig 4) from the infra-red to the deep ultra-violet with relatively high average powers. Compared to other widely tuneable sources - normally pulsed at low repetition rates, the peak power is relatively low thus avoiding non-linear effects in the molecules studied. The bandwidth of systems based on a dye laser are typically a few GHz, while a diode injected titanium sapphire system can give bandwidths below 100 MHz.

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