THE GENERATION OF TUNABLE COHERENT RADIATION IN THE WAVELENGTH RANGE 2300–3000 Å USING LITHIUM FORMATE MONOHYDRIDE*

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The generation of continuously tunable UV radiation in the wavelength range 2300–3000 Å by frequency doubling the output of a nitrogen pumped dye laser is described. A lithium formate monohydride crystal 10 mm long yields a conversion efficiency of typically 2% at fundamental powers in excess of 50 kW and allows the generation of harmonic radiation at wavelengths 150 Å below those attainable using a refrigerated ammonium dihydrogen phosphate crystal.

The production of coherent UV radiation by frequency doubling the output of a dye laser has been reported by several workers [1–5]. Ammonium dihydrogen phosphate (ADP) is commonly used to generate second harmonic radiation from fundamental wavelengths in the visible spectrum. However, whilst ADP has a high conversion efficiency it does have the disadvantage that the shortest harmonic wavelength at which phase matching may be achieved at room temperature is approximately 2620 Å. Refrigeration of the crystal allows shorter harmonic wavelengths to be generated but the lower wavelength limit, calculated at the Curie temperature of −125°C, is 2442 Å [6]. Lithium formate monohydride (LFM) has been utilised to intracavity double a cw rhodamine 6G laser to provide an ultraviolet source in the spectral range 2900–3150 Å [7]. The phase matching angles for LFM calculated from the room temperature data of Singh et al. [8] are shown in fig. 1. It is seen that phase matching can be obtained at room temperature at wavelengths shorter than in ADP. The lowest second harmonic wavelengths that may be generated using LFM are set by the absorption properties of the LFM itself which will be discussed later. In the present work the performance of LFM as a frequency doubling medium for the output of a pulsed, tunable dye laser is investigated.

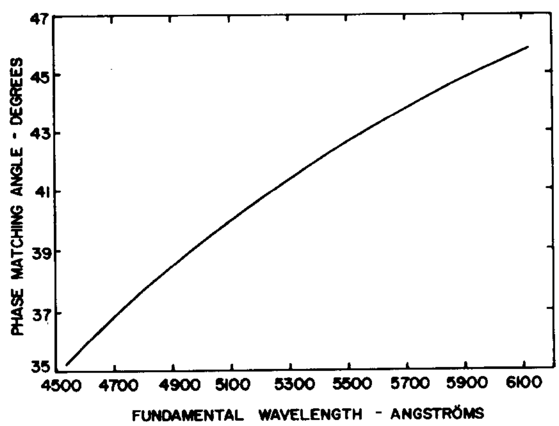

Fig. 1. Phase matching angles for lithium formate monohydride.

The nitrogen laser pumped dye laser utilised in the present study has been discussed in detail elsewhere [9] and is only briefly described here. Two new laser dyes [10] are however used in this work. The central component of the laser is a transversely pumped cell through which the dye flows. Wavelength selection is

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provided by a grating which forms one end of the cavity. A 60° prism within the laser cavity performs the dual function of beam polariser and expander. The output emerges through the uncoated fused quartz flat which serves as the end mirror of the cavity. The output beam is better than 90% horizontally polarised and has a beam divergence half angle of 2 mrad. This beam divergence results partly from the limited number of cavity transits possible during the short period for which lasing occurs, 5–8 nsec, and partly from diffraction effects associated with the small lateral dimension of the excited volume of dye. The output of the dye laser is coupled via a long focal length lens into the LFM crystal used as second harmonic generator. The LFM crystal, a cube of 10 mm side, was cut at a phase matching angle of 42°. The crystal is mounted in a sealed cell filled with index matching fluid. The end windows of the cell are broad band antireflection coated at the wavelengths of interest. The orientation of the LFM is such that the fundamental beam propagates in the XZ plane and is polarised along the Y axis. Rotation of the crystal through angles of a few degrees about the Y axis allows phase matching to be achieved over the wavelength range reported here.

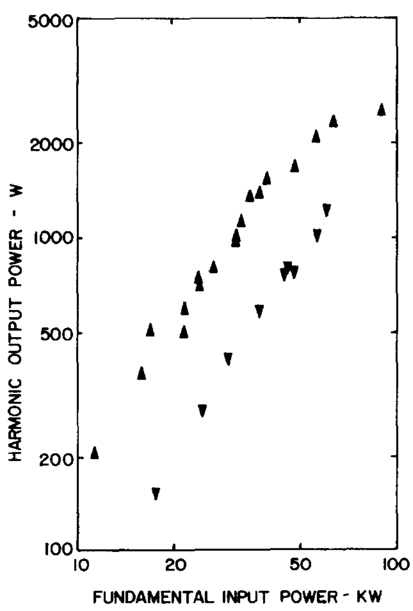

Fig. 2. The second harmonic output power as a function of the fundamental input power: ▲, fundamental wavelength 4900 Å; ▼, fundamental wavelength 6000Å.

The output of the laser is focused at the crystal in order to achieve a high power density and more efficient second harmonic conversion. The effects of focusing have been exhaustively studied theoretically [11, 12]. Boyd and Kleinman [12] show, for a gaussian beam, that as the focusing is increased the second harmonic power passes through a maximum. In the present work a 100 mm focal length lens was found to provide optimum coupling. The resulting beam convergence half angle in the crystal is 3.2 mrad.

Fig. 2 shows the power in the doubled beam as a function of the fundamental input power to the crystal at two fundamental wavelengths, 4900 and 6000 Å. At the lower input power levels the second harmonic power increases approximately as the square of the input power, a result in agreement with the theoretical predictions of Boyd and Kleinman. The observed conversion efficiency is however lower than predicted on the basis of this steady state theory and reasons for this discrepancy have been discussed elsewhere [5]. The conversion efficiency also increases with decreasing fundamental wavelength, a result again in agreement with theoretical predictions. At the higher input power levels the conversion efficiency no longer increases as the input power but tends to a constant value.

The output pulse power of the dye laser and the second harmonic power generated are shown in fig. 3. These results were all obtained at fundamental linewidths less than 2 Å. Surface reflection losses at the coupling lens and other interfaces decrease the input power to the LFM crystal by approximately 10%.

The new laser dye coumarin 102 (C102) was found to lase very efficiently over the wavelength range 4530–5120 Å under nitrogen laser excitation. The energy per output pulse is comparable to that of rhodamine 6G (Rh6G). The output pulse power from C102 is however greater than that of Rh6G because of its shorter output pulse width. An acidified solution of C102 was found to lase at a reduced output power level over the wavelength range 5090–5780 Å. Attempts to lase coumarin 6 (C6) in the present laser were unsuccessful, partly because of its low solubility in alcohol and partly because of its apparent low absorption at 3371 Å. C6 may however be successfully pumped by excitation transfer from an admixture of C102. The resultant dye mixtures lase efficiently at wavelengths up to 5350 Å and provide output powers

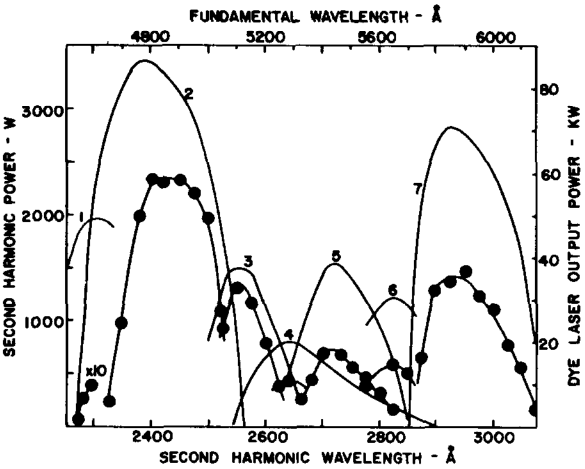

Fig. 3. The second harmonic output power as a function of wavelength: —— fundamental dye laser power; –•– second harmonic output power. 1, 7-diethylamino-4-methylcoumarin in ethanol (EtOH); 2, coumarin 102 (C102) in methanol (MeOH); 3, mixture C102 and coumarin 6 in MeOH; 4, acidified C102 in MeOH; 5, fluorescein disodium salt (FDS) in MeOH; 6, mixture FDS and rhodamine 6G (Rh6G) in EtOH; 7, Rh6G in EtOH.

in excess of those previously attained using dye mixtures based on 7-diethylamino-4-methylcoumarin [9].

The second harmonic conversion efficiency is typically 2% at input powers in excess of 50 kW. Ultraviolet beams with pulse powers in excess of 500 W have been generated throughout the wavelength range 2360–3000 Å. The conversion efficiency of an LFM crystal is however less than that of an ADP crystal of similar size and ADP would therefore appear to be a better choice of doubling material, at least for the generation of harmonic wavelengths above approximately 2620 Å. However , in order to achieve phase matching at shorter wavelengths it is necessary to refrigerate an ADP crystal, a procedure not required to obtain phase matching in LFM. Further, refrigeration of an ADP crystal to its Curie temperature will only allow the generation of harmonic wavelengths longer than approximately 2442 Å whereas fig. 3 demonstrates that LFM may be used at room temperature to generate multikilowatt pulse power UV beams at wavelengths as short as 2375 Å. The conversion efficiency and hence harmonic output power decrease rapidly below this wavelength because of the absorption of the harmonic radiation by the LFM crystal itself. Appreciable second harmonic powers may nevertheless be generated at wavelengths as low as 2300 Å. Low power harmonic radiation was observed at wavelengths as low as 2250 Å. The input power density must however be reduced when operating in this region to prevent the absorbed harmonic radiation from burning the crystal.

The present laser system was developed as a source of tunable radiation for use in the study of a variety of photon interaction processes. The use of LFM as a second harmonic generator extends the short wavelength coverage approximately 150 Å below that attainable using ADP. The range of application of the system is thereby increased and a variety of new experiments, such as a study of the photodissociation of O_2 which has a threshold at 2440 Å, may be contemplated.

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