PROPERTIES AND GROWTH OF Diamond

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7.4 Diamond lasers

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A INTRODUCTION

Many of the properties which make diamond uniquely attractive for mechanical and electronic applications also lend interest to its potential for optical applications. In particular, its wide bandgap and the exceptional stability of various optical centres in this host make it attractive for generation of radiation throughout the visible spectral region. Furthermore, the high thermal conductivity, excellent hardness, and high damage threshold of diamond suit it well to high power laser operation. Here we present a review of progress and prospects in diamond laser development.

In Section B, a few radiation-induced defect centres which may be useful for the generation of laser radiation in diamond are compared with high gain laser centres of the alkali halides. In Sections C and D, progress on H3 laser development and prospects for other diamond centres are discussed.

B OPTICAL CENTRES FOR LASERS IN DIAMOND

Pure diamond is an indirect gap material like silicon. As a consequence, there is little hope of obtaining stimulated emission directly at the bandgap recombination wavelength of 227 nm [1]. However intrinsic point defects, impurities, and centres composed of both point defects and impurities provide potential candidates for laser action at longer wavelengths. Here, we tabulate spectroscopic parameters and theoretical laser gain for a few centres which combine impurities and vacancies and review their potential for laser applications. Procedures for the synthesis of centres and general characteristics of colour centre lasers may be found in [2] and [3] respectively.

From TABLE 1 two general conclusions may be drawn. First, diamond colour centres have exceptionally broad fluorescence linewidths $\Delta\nu$ compared to those of the commercially successful alkali halide lasers, exemplified by the F_2^+ laser. As a result, their potential tuning range is many times greater. Second, their gain coefficients are lower than that of F_2^+:KCl, but still very respectable considering the breadth of emission. The order of magnitude difference between F_2^+ and N-V gain coefficients may be overcome in principle by merely increasing the excited state density by a factor of ten.

TABLE 1 Comparison of diamond and F_2^+:KCl colour centre gain coefficients γ for constant inversion density $N = 10^{16}$ cm^{-3}.

Centre	n	T (K)	τ_{fl} (ns)	η	$\Delta\nu$ (10^{13} s^{-1})	λ_o (μm)	γ (cm^{-1})
F_2^+	1.4841 [3]	77	τ_{fl}/η = 80 [3]	τ_{fl}/η = 80 [3]	1.69 [3]	1.680 [3]	3.52
N-V	2.4065 [4]	295	13.3 [5]	0.99 [5,6]	6.53 [5]	0.697 [5]	0.366
H3	2.4262 [4]	295	16 [2]	0.95 [7]	5.27 [5]	0.531 [5]	0.201

C PROGRESS ON H3 LASERS

Since the discovery of the first diamond laser [8], two other experiments have demonstrated laser action in diamond using H3 colour centres. In this unpublished work, high densities of H3 centres in natural and synthetic diamonds were used, as described briefly below.

In 1988 at the Itami Research Laboratories of Sumitomo Electric in Japan, a flashlamp-pumped Coumarin dye laser with pulsewidth of 500 ns was used to side-pump a natural diamond crystal of dimensions 3 x 2 x 2 mm^3 at an excitation wavelength of 490 nm inside a 2-mirror resonant cavity. The end mirror was a total reflector with a radius of 40 cm and the output mirror had the same curvature and a reflectivity of 97%. Both mirrors were spherical and separated by approximately 35 cm. Laser action in the yellow spectral region was obtained at room temperature with a threshold of 3 MW/cm^2 without auxiliary cooling [9]. Laser output ceased when cavity mirrors were detuned or blocked, thus extending the original result by the use of an external cavity.

Continuous-wave laser action was demonstrated in the same year at the University of Strathclyde [10]. Optical excitation was performed with a continuous-wave argon ion laser operating at 488 nm. In this case a 3-mirror cavity was used to compensate for astigmatism introduced by tilting the sample to Brewster's angle for minimum reflective losses, and the crystal was cooled to 77 K. Output power is shown as a function of input power in FIGURE 1 for two synthetic diamond samples from De Beers. Although slope efficiency was very low (6.6 x 10^{-4}%), the tuning range of this laser was remarkably wide, comparable with the fluorescence spectrum shown in FIGURE 2. Its low efficiency may be related to residual N-V absorption in the laser emission region, or to transient losses associated with B or Ni impurities used to facilitate H3 formation during crystal growth. Sumitomo researchers have also synthesised H3 crystals [11] with very bright photoluminescence, but to date have not reported continuous lasing.

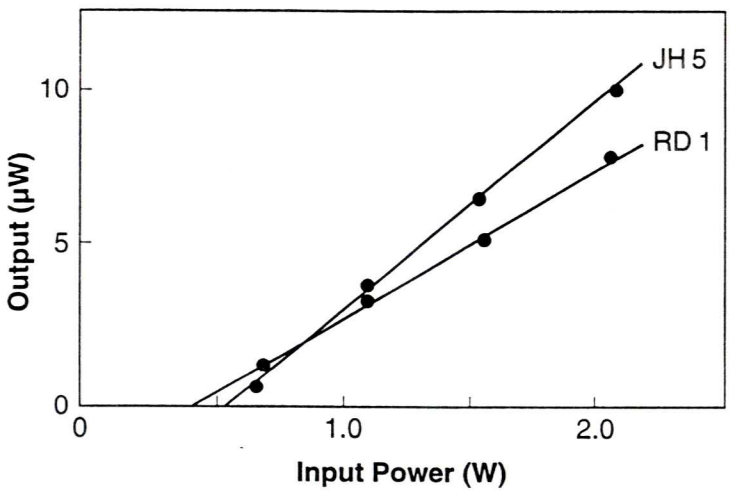

FIGURE 1 Continuous-wave laser output power as a function of input pump power at 488 nm in two synthetic diamond crystals (De Beers JH5, RD1) at T = 77 K. (With permission [10].)

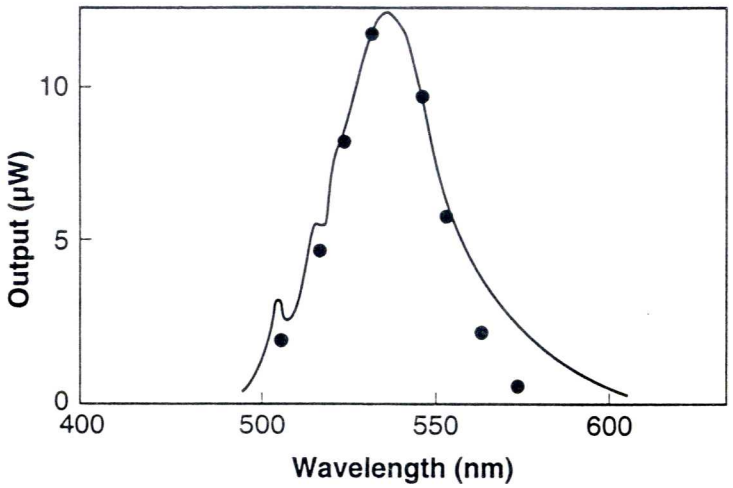

FIGURE 2 Comparison between tuning range of the CW diamond laser (solid circles) and the fluorescence spectrum (solid curve) of the laser active H3 centre, obtained by time resolved photoluminescence. (With permission [10].)

D PROSPECTS FOR NEW DIAMOND LASERS

The H3 laser described above is the only laser of any kind able to scan continuously through the wide visible range from 500 - 600 nm. This unique capability argues strongly for renewed research to improve crystal synthesis in an effort to minimise parasitic losses. Additionally however, other promising approaches for lasers at both shorter and longer wavelengths in diamond should be considered. Several of these are outlined below.

The theoretical gain coefficient for the N-V centre (TABLE 1) is nearly twice that of the H3 centre. Hence it is an excellent laser candidate, compatible with efficient diode laser pumping schemes. Of course, absorptive losses in the emission band must be kept very low for net positive gain to be realised experimentally. Encouragingly, the only centre with broadband absorption in the range 650 - 750 nm is the GR1 vacancy centre, which exhibits very low absorption (and emission) at room temperature and can be removed by appropriate annealing procedures. Also, the intrinsic quantum efficiency of the N-V centre is unity [6]. Although recent spectroscopic studies [12] confirmed the presence of a metastable level below the 638 nm fluorescent level of the N-V centre which acts as a sink for excited state population, its effect is negligible. Furthermore, high concentrations of this centre can be produced by a simple procedure, so its overall prospects for laser action remain good.

Other nitrogen-related centres may be viable laser candidates. Scant information is available on detailed models or stability of such centres, however, and few have strong broadband emission in the visible or near infrared (IR) spectral regions. Other approaches nevertheless offer promising new directions. For example, changing diamond into a direct gap material through the use of strained layer structures has been considered [13]. Additionally, radiation-induced and aggregate impurity centres in boron-doped diamond should be investigated. Boron is easily substituted into the diamond matrix and results in useful semiconductivity. By incorporating other impurities which fit reasonably well into the lattice (C^{+4} ionic radius = 0.16 Å), like nitrogen, sulphur, chlorine, phosphorus and silicon (ionic radii of 0.16 Å, 0.44 Å, 0.37 Å, 0.34 Å and 0.42 Å respectively), many new stable aggregate

centres can undoubtedly be created and pumped electrically, warranting further investigation. By combining such centres with radiation-induced vacancies using suitable annealing procedures, additional possibilities for stable centres may be realised.

Rare earth (RE) dopants, which are currently under investigation for lasers and display components in compound semiconductors [14], and constitute the laser active species in dozens of commercial solid state lasers, may also be useful for semiconductor diamond defect lasers. However, the ionic radii of trivalent rare earth ions all exceed 0.86 Å. Hence it seems unlikely that high enough concentrations of such impurities can be achieved in diamond to permit such applications. Transition metal ions tend to have smaller radii and may therefore be more promising candidates for laser research in this host. Boron itself absorbs strongly throughout the visible spectrum, so it is likely that in B-doped semiconducting diamond useful emission wavelengths of impurity metal laser ions (possibly associated with compensating, radiation-induced defects) will be restricted to the infrared. At very long wavelengths because of its renowned transparency in the far infrared (FIR), diamond might ultimately prove, however, to be the material of choice, indeed perhaps the only material compatible with solid-state FIR laser operation.

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[6] Optical pumping losses from the lowest ^{3}E level (labelled 4) to both the ^{1}A state (3) and the second (spin-split) ground state level (2) estimated from unpublished transient hole-burning experiments together with results of [12,15]. Absolute changes in population of metastable levels were determined and compared with steady-state theoretical population ratios governed by simple decay ratios. Values determined in this way were $\gamma_{42} = 2.2 \times 10^4\ s^{-1}$ and $\gamma_{43} = 7.8 \times 10^4\ s^{-1}$. Since $\gamma_4 = (\pi \times 13.3 \times 10^{-9})^{-1}$ [5], we find intrinsic quantum efficiency $Q = \gamma_{41}/(\gamma_{41} + \gamma_{42} + \gamma_{43}) = 0.99$.

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