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Year 2023	**Summary of Thesis**
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(Title)
Laser Oscillation in DNA/CTMA/Fluorescent Dye/Azo Dye Complex

In recent years, DNA dye complexes have been attracting attention as optical and electronic device materials, and many studies on the application of bio-derived materials to optical and electronic devices have been reported. Among them, simple and compact wavelength-tunable lasers, which are difficult to realize with current technology, are expected to be applied in various fields such as clinical medicine and environmental measurement. We have previously reported a feedback-distributed laser oscillation based on the formation of dynamic gratings by the injection of two fluxes of excitation laser light into a DNA/CTMA/cyanine dye complex, utilizing the fact that doping a cyanine dye into a complex of DNA and a quaternary ammonium salt called CTMA with lipid increases the weak fluorescence. Furthermore, utilizing the periodic structure of the refractive index due to the photoisomerization reaction of azobenzene, we have developed a two-layer feedback-distributed laser with a DFB layer of PMMA doped with azo dye (DO3/DR1) and confirmed laser oscillation (Figure 1). However, this double-layer laser can only oscillate for 10 minutes (short lifetime) and has a high oscillation threshold (0.1~1.0 mJ/cm^2).

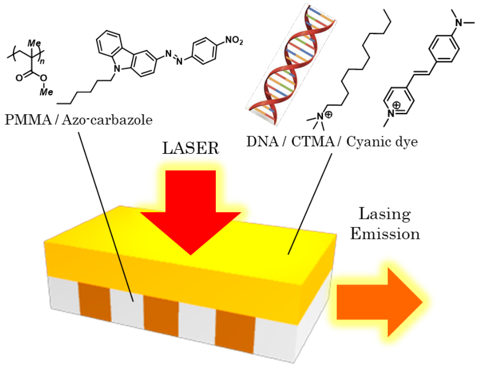

Figure 1. Monolithic DNA complex dye laser.

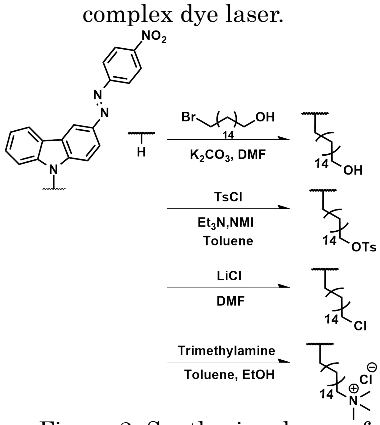

Figure 2. Synthesis scheme of Azo dye-CTMA

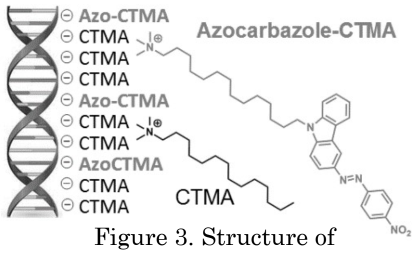

Figure 3. Structure of DNA/CTMA/Azo dye complex.

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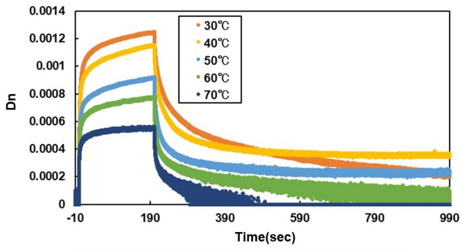
In this study, we collaborated with Kawabe's laboratory to realize a simple compact wavelength-tunable laser with low threshold and long life by simultaneously creating luminescence by cyanine dye and periodic structure of refractive index by photoisomerization reaction of azo dye on a DNA/CTMA template. Azo-carbazole was used as azo dye, and modified with a long-chain alkyl quaternary ammonium salt (azo-carbazole -CTMA, Figure 2) to prepare a ternary complex of azo-carbazole-CTMA/CTMA/DNA (Figure 3). We evaluated the temperature dependence of the birefringence relaxation of this composite by measuring the transmitted light intensity of the writing laser and the reusability of the samples by thermal refreshment. The effect of chemical modification of DR1 with CTMA (Disperse Red 1) on the birefringence change was also investigated to elucidate the mechanism: samples were prepared by simply mixing DR1 with DNA / CTMA, and the results of transmitted light intensity measurements and circular dichroism measurements were compared with those of DNA/CTMA/DR1-CTMA.



	30℃	40℃	50℃	60℃	70℃
τ_1	19.759	14.291	10.867	8.7537	4.7485
τ_2	202.54	101.46	127.83	186.22	153.67

Figure 4. Temperature dependence of birefringence relaxation of Azo-carbazole-CTMA/CTMA/DNA complex.

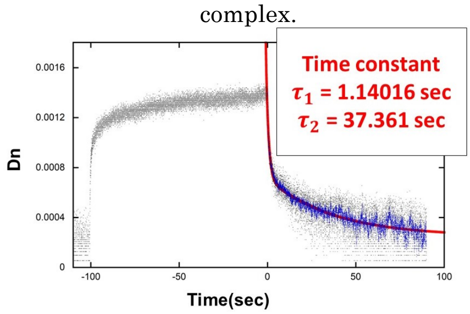


Figure 5. Relaxation of excitation birefringence of DR1 complex

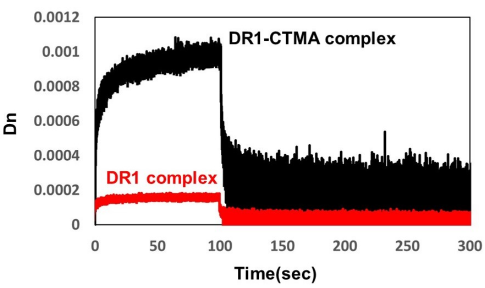


Figure 6. Relaxation of excitation birefringence of DR1 complex and DR1-CTMA complex

Azo-carbazole-CTMA/CTMA/DNA thin films were prepared and measured for transmitted light intensity. The fast and slow relaxation of birefringence were observed, and the fast relaxation time tended to decrease with increasing temperature, while the slow relaxation was not temperature dependent (Figure 4). For the DR1-CTMA composite, the birefringence of the thin films prepared by simply mixing DR1 with DNA/CTMA showed a smaller change in birefringence than that of the DR1-CTMA/CTMA/DNA complex, indicating that the chemical modification of CTMA enhances the photoisomerization of DR1 in DNA (Figure 6). Figure 6). On the other hand, DR1-CTMA showed significantly smaller birefringence change under high humidity conditions, accompanied by a peak shift of absorption wavelength to shorter wavelengths and the appearance of circular dichroism. The mechanism of these results is yet to be clarified.

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