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Citation: Applied Physics Letters 105, 011112 (2014); doi: 10.1063/1.4889080
View online: http://dx.doi.org/10.1063/1.4889080
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/105/1?ver=pdfcov
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Random lasing from dye-gold nanoparticles in polymer films: Enhanced gain at the surface-plasmon-resonance wavelength
Ultraviolet random lasing from a diamond nanoparticle film

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(Received 16 April 2014; accepted 27 June 2014; published online 8 July 2014)

We demonstrated ultraviolet random lasing from a diamond nanoparticle film, in which commercially available diamond nanoparticles with an average size of 25 nm acted as the gain medium and scatterers. By optically exciting the diamond nanoparticle film at an excitation wavelength of 355 nm, discrete sharp peaks, superposed on a collapsed broad emission spectrum, randomly appeared at wavelengths around the broad emission maximum (~382 nm) when the excitation intensity exceeded the threshold. From the dependence of lasing peak intensities measured at different locations as a function of excitation intensity, the threshold was estimated to be approximately several tens of MW/cm².

Diamond is an attractive material for applications as light sources and semiconductor power devices because of its excellent physical properties, i.e., high thermal conductivity, high carrier mobility, various color centers, and high breakdown field strength. Because diamond has a wide band gap (5.48 eV) and is transparent with a high refractive index, it can be used as a deep ultraviolet (DUV)/ultraviolet (UV) light source for sterilization and cleaning, medical applications, high density recording, and a high color-index, it can be used as a deep ultraviolet (DUV)/ultraviolet (UV) light source for sterilization and cleaning, medical applications, high density recording, and a high color-

![Image](http://dx.doi.org/10.1063/1.4889080)

FIG. 1. Emission spectra at different spots within a diamond nanoparticle film. Insets show CCD images of emissions. The excitation intensity was kept constant, and the white bars indicate the scale of 50 μm.

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wavelengths around 380 nm. Figure 2(b) shows the logarithmic plots of the peak intensity vs. the excitation intensity. From the results, we confirmed the nonlinear evolution of the emission peak intensity when the excitation intensity exceeded the threshold (about 60 MW/cm²). In addition, the observed relative peak intensities changed depending on the excitation intensity and each shot of the excitation pulse. We have to note that because diamond nanoparticles have a mean diameter of 25 nm, it is too small compared to the excitation and emission wavelengths. Therefore, it would be strange that we observed random lasing with discrete sharp peaks from the diamond nanoparticle film, because the scattering strength of individual nanoparticles should be so small to derive the random coherent feedback. To clarify this, we measured a scanning electron microscope (SEM) image and also coherent backscattering of diamond nanoparticles. The inset in Fig. 2(b) shows a SEM image of the sample. From the image, we found that the aggregation of diamond nanoparticles occurred and the film had a rough structure, which scale was enough larger than the wavelengths. In addition, from the measurement of coherent backscattering, the mean free path of the diamond nanoparticle film was roughly estimated to be about 11 times of the wavelength. Thus, we thought that the particle aggregation would play an important role for inducing random laser in the diamond nanoparticle film.

We observed the same phenomena for different sample sites and different films. Figure 3 shows the emission spectra for 4 different sites on the film. The peaks were always at ~380 nm. Figure 4 shows the scatter plot of lasing peak wavelengths and thresholds for different sample sites and different films (circles), and the gray line indicates the spontaneous emission spectrum from a blue spot in the diamond nanoparticle film for excitation below the threshold. The discrete sharp peaks clearly concentrated around the peak wavelength of the spontaneous emission, whereas their thresholds were dispersed from several tens to several hundred MW/cm². These results suggested that because of randomness, laser oscillation initiated at a wavelength with strong gain. The difference in the thresholds reflected the difference in the surrounding conditions at each location. Because these behaviors were similar to those of conventional random lasers, we concluded that UV random lasing was induced in these diamond nanoparticle films.

As a laser device using diamonds, many studies of Raman lasers have mainly been reported. However, because our observed lasing peaks were different from the typical reported Raman peaks (1333 cm⁻¹ and 1600 cm⁻¹), and different wavelengths were observed at different observation spots, we thought that Raman lasing was not the origin of the nonlinear evolution of the observed sharp peaks. Gorelik and Rakhmatullaev have reported fluorescence around 380 nm from diamond powder with a spectral shape similar to our emission spectra when the excitation intensity is below threshold; they attributed this fluorescence to the N2 defect. Furthermore, from previous studies in which the sp² layer on the surface of diamond nanoparticles suppressed emission from the diamonds, we thought that the surface conditions of diamond nanoparticles would play an important role in UV random lasing. In fact, by comparing the nanoparticle film before and after sintering, intense blue spots and UV lasing were rarely observed before sintering,
whereas they were easily observed after sintering. In addition, we thought that because of the removal of sp² layers due to annealing, surface carbon layers could not be the origin of the UV emission. On the basis of these results, we conjectured that before sintering, lasing is less likely because of emission suppression by the sp² layers on the diamond surface. Whereas after sintering, N2 defect emission would increase because of the removal of the sp² layer, resulting in an increase in UV random lasing. Therefore, we expect that optimization of the temperature and ambient nitrogen concentration during annealing could increase the probability of UV random lasing in a diamond nanoparticle film.

In summary, we demonstrated random lasing from a diamond nanoparticle film. We observed nonlinear evolution of discrete sharp peaks at wavelengths around 380 nm, a wavelength never before observed for diamond laser oscillation. Even though further investigation is necessary to understand the mechanism for the observed random lasing, considering the excellent properties of diamonds, our demonstration of random laser oscillation in diamond nanoparticle films in the UV region (λ/24 = 380 nm) suggests excellent promise for future diamond-based devices.

This work was supported by JSPS KAKENHI Grant Nos. 22681011 and 24651111.