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Title	Magnetic response of random lasing modes in a ZnO nanoparticle film deposited on a NiFe thin film
Author(s)	Fujiwara, Hideki; Kaiju, Hideo; Nishii, Junji et al.
Citation	Applied Physics Letters, 113(13), 131108 https://doi.org/10.1063/1.5040720
Issue Date	2018-09-24
Doc URL	https://hdl.handle.net/2115/75495
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Type	journal article
File Information	1.5040720.pdf

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Citation: Appl. Phys. Lett. **113**, 131108 (2018); doi: 10.1063/1.5040720
View online: https://doi.org/10.1063/1.5040720
View Table of Contents: http://aip.scitation.org/toc/apl/113/13
Published by the American Institute of Physics

Magnetic response of random lasing modes in a ZnO nanoparticle film deposited on a NiFe thin film

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(Received 20 May 2018; accepted 14 September 2018; published online 28 September 2018)

This study experimentally demonstrates lasing mode switching within a ZnO nanoparticle film coated onto a magnetic thin film of NiFe alloy. When a neodymium magnet is brought close to or moved away from the film, switching behavior is observed in the lasing modes, although such change is not induced in a ZnO nanoparticle film on a glass substrate. Our results suggest that the observed changes in lasing modes are because of a magneto-optical effect at the surface of the NiFe thin film. The magneto-optical effect would be enhanced by localized fields near the surface, inducing suppression or enhancement of the lasing modes in response to the surrounding environments, and accounting for the lasing mode switching. *Published by AIP Publishing.*
https://doi.org/10.1063/1.5040720

Wavelength-disordered structures and their localized mode control have been attracting increasing attention for use in light-emitting devices,[1–3] sensors,[4,5] and light-harvesting applications.[6–8] Although, in such applications, multiple light scattering results in random feedback, this also presents challenges in the control of localized mode properties, optimization of light input–output, electrode formation, and electrical pumping. When used as cavity structures, light-matter interaction must be improved by their modal control, while retaining the merits of simple and low-cost fabrication. A wide range of approaches to the control of localized modes have been proposed, with the goal of matching the resonant property of the material, whereas, no fully effective solution has yet emerged. These approaches have been of two main kinds. In the first approach, the resonance characteristics are controlled during fabrication (pre-processing). This allows the wavelength and threshold to be roughly controlled by adjusting the structural conditions such as the scatterer size[9–12] or absorption properties.[13–15] In the second approach, external stimuli are used, in which the lasing modes are tuned using photonic,[16,17] electric,[18,19] thermal,[20,21] or mechanical[22–24] responses.

In our previous studies, we have also proposed a number of methods for fabricating unique random structures and controlling their localized modes. To improve their lasing properties, we applied resonant scatterers (ZnO spherical nanoparticles and size-controlled nanorod arrays) fabricated using laser-melting and laser-induced hydrothermal synthesis.[11,12] Furthermore, we also demonstrated a post-processing technique for tuning the lasing modes, in which photothermal conversion and negative thermal expansion of graphene flakes took place within a ZnO nanoparticle film and an external white light source induced the modulation of random lasing modes.[24] Combining these pre- and post-processing treatments would allow the random lasing modes to be controlled, making it possible to incorporate an active tuning mechanism into the resonance-controlled random structures. This should make the randomly determined localized modes freely manipulable, in the same way as conventional cavity modes, while retaining the advantages of random structures. However, under active tuning of lasing modes using graphene flakes, the thermal response was slow (several seconds) and the strong light absorption of the graphene made the thresholds several times larger.

In this study, we investigated an alternative method for addressing these drawbacks and realizing remote, non-invasive, and cost-effective control of the random lasing modes. Our approach made use of an external magnetic field to induce lasing mode tuning or switching in a zinc oxide nanoparticle film coated onto a magnetic thin film. When localized modes form closed loops through reflection at the magnetic film layer, Kerr rotation or refractive index changes are expected to induce a change in the polarization of the photons fed back into the nanoparticle film. This should trigger changes in the lasing modes. Our experimental results demonstrated a magnetic response by random lasers in a ZnO nanoparticle film coated onto a NiFe alloy thin film. Clear changes were observed in the peak intensity of lasing as the magnetic field was applied to and removed from the sample. In Ref. 25, Tsai *et al.* recently reported a similar magnetic response in a random laser composed of TiO_2 and Fe_3O_4 nanoparticles dispersed in a dye solution. When an external magnetic field was applied, magnetic nanoparticles collected at the cuvette surface and the density of the scatterers increased and the resulting change in transport mean free paths produced a change in the lasing mode. Our present study used a different approach, in which the magnetic response of a random laser was observed in a solid phase rather than in a solution. The utilization of the magneto-optical effect in a magnetic thin film was expected to realize fast and reproducible responses. This method could allow remote, non-invasive control of random lasing modes. Considering its use of magneto-optical effect with intracavity enhancement, the approach will be applicable to conventional microcavity systems, including reflection at a magnetic thin film. Because of recent advances in the coherent coupling of photons with magnetic materials, such as optomagnonic whispering gallery microresonators[26,27] and giant Faraday rotation in magnetic nanogranular films,[28] our

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approach may contribute to the field of spintronics, as well as photonics.

A NiFe alloy thin film with a thickness of 35 nm was deposited onto a $10 \times 10 \times 0.5$-mm glass substrate using ion beam sputtering [Fig. 1(a)].[29] The base pressure before sputtering was 1.0×10^{-5} Pa, and the growth rate of the NiFe alloy thin film was 3 nm/min at a sputtering power of 60 W, under an Ar pressure of 3.0×10^{-2} Pa. The Kerr rotation of the NiFe alloy thin film was detected from focused magneto-optical Kerr effect (MOKE) measurements at a laser wavelength of 405 nm. A magnetic field of up to 10 kOe was applied perpendicular to the plane of the film at room temperature. Figure 1(b) shows the magneto-optical Kerr rotation of the NiFe film. When the magnetic field strength is less than 9 kOe, the Kerr rotation is proportional to the field, but becomes saturated at 200 mdeg above 9 kOe. Commercially available ZnO nanoparticles (diameter, ~100 nm) were dispersed in water and used as scatterers and gain materials. A drop of the solution was cast on the NiFe film and dried at room temperature [Fig. 1(c)]. As a control, a ZnO nanoparticle film was prepared in the same way on a glass plate.

The sample was set on a home-made aluminum microscope stage and irradiated with pulses from a Q-switched pulsed laser (wavelength, 355 nm; repetition rate, 1 kHz; pulse duration, 300 ps; Teem photonics) focused by an objective lens (60×, NA = 0.85, spot size ~60 μm) as the excitation light. Emission from the sample passing through a pinhole, for which an image size on the sample surface was ~1.7 μm, was guided to a spectroscope equipped with a cooled CCD camera. An external magnetic field was applied by a neodymium magnet placed 1 mm from the NiFe film. This provided a field strength of ~5 kOe at the surface of the NiFe film. To observe the effect that the magnetic field had on the random lasing properties, we measured the emission spectra from the ZnO nanoparticle film while changing the excitation intensity.

Figure 2 shows the excitation intensity dependence of emission spectra as the magnetic field was applied and removed. While some peaks were affected by the field [an example is marked by a circle in Fig. 2(a)], one peak was not

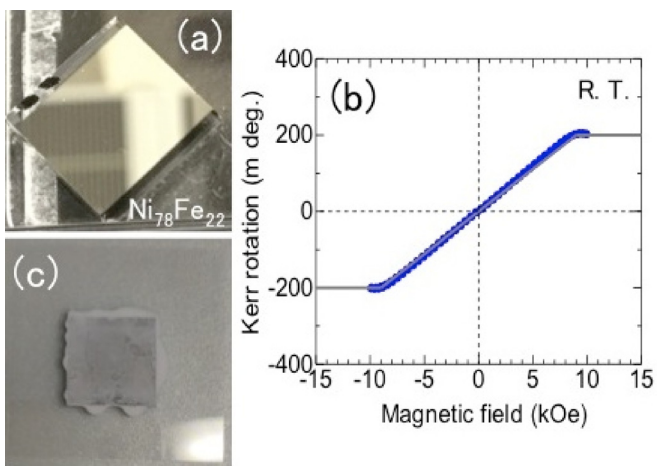

FIG. 1. (a) Image and (b) Kerr rotation of the NiFe thin film (35 nm). A magnetic field was applied perpendicular to the film plane, and Kerr rotation was detected from laser light reflection at 405 nm. (c) Image of the ZnO nanoparticle film cast on the NiFe film.

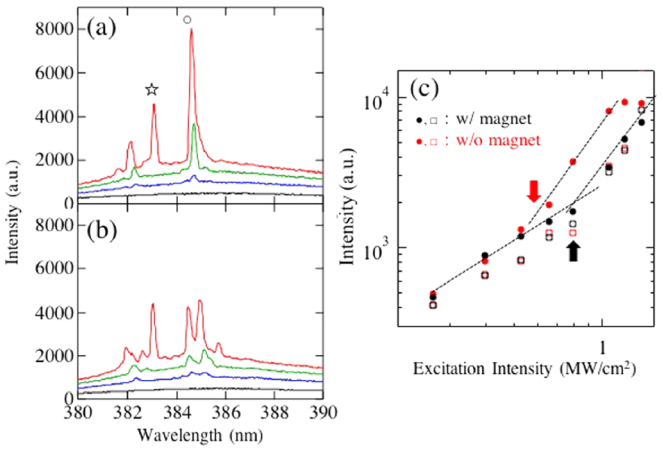

FIG. 2. Excitation intensity dependence of emission spectra in the (a) absence and (b) presence of the magnetic field. The excitation intensities were 0.26, 0.53, 0.79, and 1.05 MW/cm^2 from bottom to top in each figure. (c) Excitation intensity dependence of emission peak intensities at 384.7 and 383.0 nm (indicated by circles and squares, respectively) with and without a magnetic field. These peaks correspond to the peaks indicated by a circle and star in Fig. 2(a).

responsive [marked by a star in Fig. 2(a)]. Figure 2(c) shows the emission peak intensities at 384.7 and 383.0 nm [marked by a circle and star in Fig. 2(a)] against the excitation intensity. This confirmed a difference between the thresholds of the peak at 384.7 nm [circles in Fig. 2(c)] in the presence and absence of the magnetic field (~0.8 and 0.5 MW/cm^2). Above the thresholds, a clear difference in the peak intensity was observed, whereas no change was seen in the spectra below the thresholds. In contrast, the peak at 383.0 nm [squares in Fig. 2(c)] did not show any change in the emission intensities and thresholds between the cases with and without the magnetic field. Furthermore, we note that when the excitation intensity dependence of the integrated intensity (not shown) was plotted, no clear difference was observed between cases with and without the magnetic field. This was attributed to the changes being masked by the integration of the spectrum.

To more closely examine the influence of the magnetic field when the excitation intensities were fixed, repeated measurements were taken of the emission spectra in the presence and absence of the magnetic field (Fig. 3). Figure 3(a) confirms that the spectral shape changed in a reproducible way when spectra with and without the magnetic field were superimposed. Figure 3(b) plots the change in emission peak intensities at 384.7 nm [marked by a circle in Fig. 2(a)] when the magnetic field was applied at different excitation intensities. For comparison, we also plotted the peak intensities at 382.8 nm [marked by a star in Fig. 2(a)]. The peak intensities at 382.8 nm (blue circles) showed no change in response to the magnetic field, whereas those at 384.7 nm (black circles) showed a clear dependence: when the magnetic field was applied, lasing was suppressed. This phenomenon was not observed in the ZnO nanoparticle film cast on a glass substrate. Note that the peak intensities at 382.8 nm are plotted only in the upper part of Fig. 3(b), as lasing was already suppressed when the excitation intensity was less than 0.79 MW/cm^2, and the intensity of fluorescence showed no change against the magnetic field.

Similar magnetic responses can also be observed at different positions and samples. Figure 4 shows the results

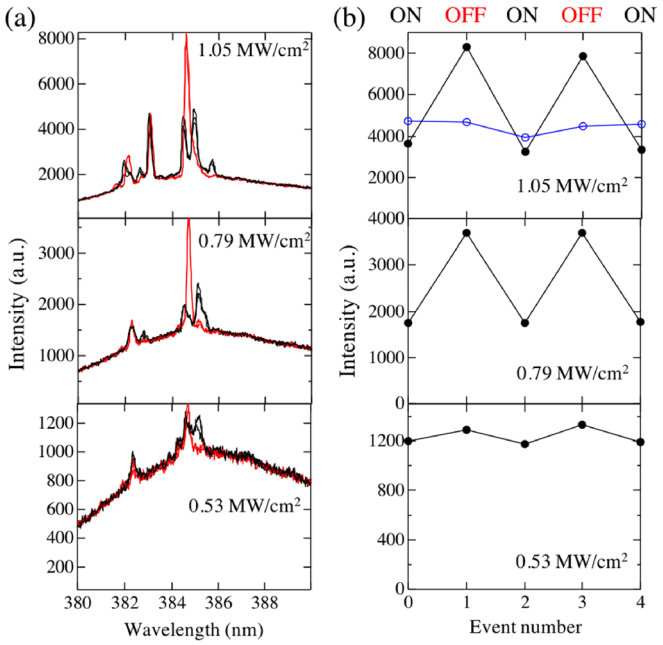

FIG. 3. Magnetic field dependence of (a) emission spectra and (b) peak intensities. The excitation intensities were fixed at 1.05, 0.79, and 0.53 MW/cm^2. The black and red lines in (a) indicate spectra in the presence and absence of the magnetic field. These are the same data shown in Figs. 2(a) and 2(b). In (b), the black circles indicate the peak intensities at 384.7 nm and the blue circles indicate the peak intensities at 382.8 nm [marked by the circle and star in Fig. 2(a)].

when measured at different positions. Random lasing and a magnetic field response were also observed. In these measurements, the magnetic field was applied and removed five times. When all data were superimposed, as shown in Fig. 4(a), the emission spectra corresponded closely with each other, confirming the reproducibility of this response. Figure 4(b) plots the emission intensities of three different peaks [marked by the arrows in Fig. 4(a)]. Again, the peak intensities changed in response to the magnetic field. However, in contrast with the results shown in Fig. 3, both suppression (black and red circles) and enhancement (blue circles) of random lasing were observed in response to the magnetic field. As the lasing modes were shown to be suppressed, promoted, or unaffected by the magnetic fields, we considered that no change in absorption of the NiFe film had taken place. Instead, the changes were attributed to the formation of closed loops in the lasing mode, due to reflection at the NiFe

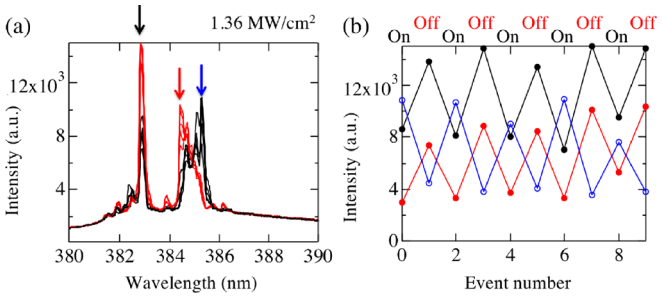

FIG. 4. Magnetic field response of (a) emission spectra and (b) peak intensities. The excitation intensity was fixed at 1.36 MW/cm^2. The black and red lines in (a) indicate the spectra with and without the magnetic field. The black, red, and blue circles in (b) denote the peak intensities, indicated by arrows in (a).

film surface. We conjectured that Kerr rotation or the change in the refractive index of a NiFe thin film is one of the possible mechanisms for the magnetic response of random lasing by applying the magnetic field, by which the feedback of the closed loop closed to the surface may change the lasing modes. Although Kerr rotation from the application of 5 kOe perpendicular magnetization would be only of the order of 100 mdeg, we therefore conjecture that an increase in the influence of Kerr rotation from the intra-cavity effect, and a gain change produced by feedback to the mode, produced the steep change in lasing peak intensities. Similarly, in the case of the refractive index change, it would also induce lasing intensity change and/or switching the lasing modes, for which modes were located closed to the NiFe surface.

Figure 5 shows the dependence of the emission spectra and peak intensities on the magnetic field intensity. In order to measure the magnetic field intensity dependence, the neodymium magnet was fixed on a motorized stage, and the distance between the magnet and sample was controlled to change the magnetic field intensity applied to the sample while the excitation laser intensity was kept constant (1.15 MW/cm^2). In the experiments, we measured emission spectra with changing the magnetic field intensity, in which the magnetic field was increased from 0 to 5500 Oe, then decreased to 0 Oe, and increased again to 5500 Oe to confirm the reproducibility. Figure 5(a) shows the emission spectra measured when the magnetic field intensity was increased (0, 2900, and 5300 Oe). At the lasing peaks (b) and (c), the peak intensity changed depending on the presence or absence of the magnetic field as shown in the previous results, while almost no change in the peak (d) against the magnetic field was observed. However, from the magnetic field intensity dependence in Fig. 5(d), although almost no change in the emission peak intensity at the minimum and maximum magnetic field intensities was confirmed, we found that when the magnetic field intensity decreased, the peak intensities show different states from

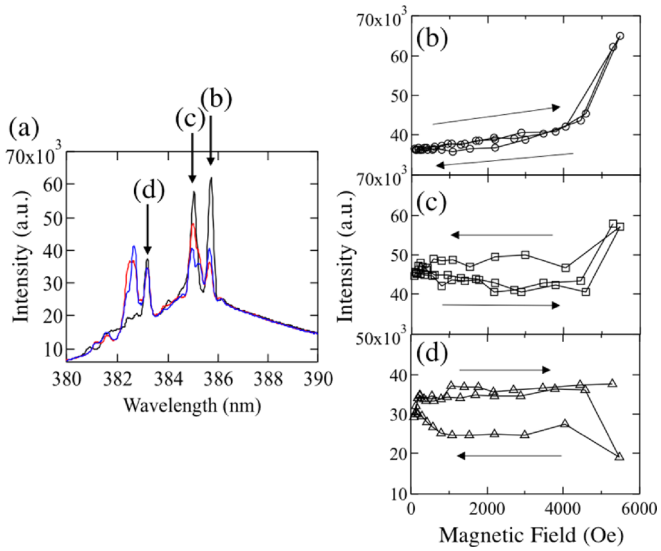

FIG. 5. Magnetic field dependence of (a) emission spectra and (b)–(d) peak intensities when the excitation intensity was fixed at 1.15 MW/cm^2. The red, blue, and black lines in (a) indicate the emission spectra with a magnetic field of 0, 2900, and 5300 Oe, respectively. The circles, squares, and triangles in (b)–(d) denote the peak intensities indicated by arrows in (a). The arrows in (b)–(d) indicate the directions of magnetic field intensity change.

those when the magnetic field intensity increased, which behaved like bistability. On the other hand, at the peak (b), the steep emission intensity change can be observed from around 4000 Oe, and the peak (c) behaves like an intermediate state between the behaviors between the peaks (b) and (d). These results suggested the possibility that the change depending on the magnetic field intensity was induced even at the peaks where no change was observed in the On-Off measurements. Threshold- and bistability-like behaviors against the magnetic field intensity [Fig. 5(b)] could be explained that the small change in random feedback due to the magneto-optical effect of a magnetic substrate would be enhanced by the optical confinement and amplification process. As a result, the change in the lasing property at a specific magnetic field intensity would be induced. However, in order to elucidate the mechanism, although the observation of phenomena induced at the vicinity of a magnetic thin film is indispensable, the experimental verification is difficult due to the strong light scattering. For this purpose, we consider that numerical approach is necessary for understanding the mechanism as a future work.

In summary, we demonstrated the modulation of random lasing modes in a ZnO nanoparticle film coated onto a NiFe alloy thin film when an external magnetic field was applied. The lasing spectral response to the presence and absence of the magnetic field suggested that the changes in lasing modes would be attributed to the magnetic response of the NiFe thin film. Our method may provide an approach to the remote, non-invasive control and tuning of random lasing modes, in which a magnetic field provides the external stimulus, and also be applicable to other conventional microcavity systems. Although the current study used a random structure, recent advances in the coherent coupling of spintronics with microcavity systems[26,27] suggest that this approach may open insights into spintronics, as well as photonics.

This work was supported by JSPS KAKENHI (Grant Nos. 17K05016, 17K19019, and 16H06506), Dynamic Alliance for Open Innovation Bridging Human, Environment and Materials, the Amada Foundation, and Nippon Sheet Glass Foundation for Materials Science and Engineering.

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