Research article

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Flexibly tunable surface plasmon resonance by strong mode coupling using a random metal nanohemisphere on mirror

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Abstract: We propose a unique random metal nanohemisphere on mirror (NHoM) structure to tune the surface plasmon (SP) resonance in a flexible manner. The SP resonance peak was split into two peaks owing to the strong coupling between the SP mode in the metal nanohemisphere and the mirror image mode generated in the metal substrate. This phenomenon is based on the fact that the strong coupling and the induced electromagnetic effects are similar to those pertaining to the Rabi splitting, Fano resonance, and electromagnetically induced transparency, thus providing quantum effect analogies. These phenomena have recently attracted increased attention and have been studied with nanocavities fabricated with top-down nanotechnologies. Compared with previous reports, NHoM structures can be fabricated in a much easier manner and are tunable in rather wider wavelength regions without nanofabrication technologies. The SP resonance peaks were enhanced, sharpened dramatically, and tuned flexibly, based on the optimization of the thickness of the spacer layer between the metal hemisphere and metal substrate. Experimental results were reproduced and were explained based on finite difference time domain (FDTD) simulations. These phenomena have never been observed previously on similar nanosphere on mirror (NSoM) because nanohemispherical structures were required. The NHoM nanocavity structure has a quality factor >200 that is surprisingly high for the localized SP

mode of nanoparticles. Flexible tuning of the SP resonance with the use of NHoM is envisaged to lead to the development of new applications and technologies in the field of plasmonics and nanophotonics.

Keywords: mode coupling; nanohemisphere on mirror; plasmonics; strong coupling; surface plasmon.

1 Introduction

Plasmonics enable the manipulation of light waves at the nanoscale—that is, at a much smaller scale than the wavelength scale—by inducing resonance with surface plasmons (SP) generated at the metal/dielectric interface [1]. Following the remarkable developments of nanotechnologies in recent years, plasmonics has been expected to become a fundamental and platform technology that will lead to various new classes of nano-optical applications. Various new optical nanotechnologies operating below the wavelength size have been proposed, including plasmonic bandgap structures [2], plasmonic waveguides [3], and anomalous light transmission using nanohole arrays [4]. In 2001, our project team at the California Institute of Technology in collaboration with Atwater have introduced the term “plasmonics” for new research fields dealing with light and its interactions at the nanoscale via SP [5]. Plasmonics has been expected to lead to the development of various new fundamental technologies, such as nano-optical circuits and computing, that will support the future ultrasmart society [6]. For example, we succeeded in increasing the efficiency of light-emitting materials by plasmonics for the first time in 2004, and proposed that these can be applied to high-efficiency light-emitting devices [7]. This method is expected to be used in device application and has been studied by many research groups [8]. Recently, we successfully fabricated the prototype of the high-efficiency plasmonic light-emitting diode (LED) [9]. A similar mechanism has also been used to improve the efficiencies of solar cells [10].

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SP resonance is also a type of nano-optical resonator, and high-quality factors (Q) and nanoscaled mode volumes (V) can be achieved by creating special and complicated structures using top-down microfabrication technologies [11]. In recent years, nanolasers based on the SP resonance with propagating mode with semiconductor nanorods [12, 13], or localized mode with nanoparticles [14, 15], have been reported and have attracted increased attention. In the field of cavity quantum electrodynamics (QED), various electromagnetic effects similar to quantum effects have been observed, such as normal mode splitting [16, 17], electromagnetically induced transparency [18–20], and Fano resonance [21, 22]. Rabi splitting [23–25] and the Purcell effect [26–28] have also been respectively observed based on strong and weak couplings generated between the excited state and the cavity mode. Moreover, the flexible control of the optical properties, including negative refractive indices and novel phenomena, such as a complete absorption/reflector or slow light, have also been studied based on the plasmonic metamaterials [29].

In recent years, however, these research fields have become saturated, and leaps and bounds will probably not be expected in the future. Additionally, electron beam lithography, ultrahigh vacuum drying processes, and others, have improved the accuracy of nanofabrication processes, and the sizes of these electronic devices are approaching the limit of Moore's law. However, these processes are not so suitable for industrial applications owing to the large cost and time required to process large areas. Various nanostructures, such as dolmen [21], double-ring [30], split-ring structures [31], and chiral characters [32], and novel optical properties based on them have been studied, but it is difficult to apply them to large areas. Furthermore, many structures are fabricated on the material surface in two-dimensions. We believe it is very important and required to establish a new method to fabricate nanostructures easily and inexpensively in large areas, and three-dimensional structures based on bottom-up processes. These techniques and nanostructures will lead to the development of a variety of optical technologies and devices, and will become useful in a wide range of fields.

The use of metal nanoparticle array structures to control localized SP modes is a promising method. Quite recently, we found unusual optical properties of three-dimensional multilayers of silver (Ag) nanosheet located on metal substrates [33, 34]. The peaks of the extinction spectra were split into two peaks, but this phenomenon was not observed on a transparent substrate. This optical phenomenon is due to the mode-splitting effect attributed to the strong coupling. Accordingly, the Ag sheet structures

can act as plasmonic metamaterials. By using this structure, we reported several applications, such as colorimetric biosensing [35], photocatalytic reaction sensing [36], enhanced fluorescence imaging [37], and ultrahigh resolution imaging [38]. Herein, we introduce a new plasmonic metal nanostructure named nanohemisphere on mirror (NHoM) that provides similar splitting, enhancement, and increased Q -values of the SP resonance by similar mode coupling through the mirror image even with the use of random structures that are much simpler and easier to fabricate. Furthermore, by simply changing the thickness of the spacer layer, flexible tuning of the mode coupling condition and resonance peak position in wider wavelength ranges can be achieved without changing the shape and size of the nanoparticle structure.

2 Methods

The samples of the NHoM structures were fabricated as follows: Ag layers with thicknesses of 100 nm were deposited on BK7 glass substrates by a thermal evaporator, and silicon dioxide (SiO_2) layer spacers were deposited by a radiofrequency (RF) sputter at various thickness values (5–20 nm). Thin Ag layers (thickness of 10 nm) were deposited on the spacer layers, and were thermally annealed with 200 °C in an electric furnace with a nitrogen atmosphere to form Ag nanoparticle (NP) structures. For the reference sample, a Ag nanohemisphere on glass (NHoG) structure was also fabricated on BK7 glass substrates with similar methodologies. These fabrication processes are shown in Figure 1(a).

The surface morphology was investigated by high-resolution scanning electron microscopy (SEM) (Hitachi High-Technologies SU8000, Japan). The reflection spectra were monitored via an ultraviolet-visible (UV-vis) spectrometer with a reflectance measurement attachment module (5° incident angle, Shimadzu UV-1800, Japan), and were converted to extinction spectra. The light extinction (E) of the samples on the transparent substrates were converted as $E = -\log_{10}(T/T_0)$ where T and T_0 are the transmittances of the sample and substrate, respectively. The transmitted light should extinct through absorption and scattering because the detected reflection from the sample was negligibly low. On the other hand, E of samples on the metal substrate were converted as $E = -\log_{10}(R/R_0)$ where R and R_0 are the reflectance of the sample and substrate, respectively.

Finite difference time domain (FDTD) simulations were conducted with commercial software (Poynting for Optics, Fujitsu, Japan). The periodic boundary condition was set in the X and Y directions, and an absorbing boundary condition was set in the Z direction. The model sizes for FDTD calculation with periodic structures were set to twice the diameter. A pulsed light composed of a differential Gaussian function with a pulse width of 0.5 fs was used for excitation. The peak position on the spectrum of the excitation pulse was approximately 600 THz (500 nm wavelength). The dielectric function of Ag was approximated by the Drude formula based on values reported by Johnson and Christy [39]. The refractive index of the glass was set at 1.5 without dispersion. A nonuniform mesh was used with a grid size of 1 to 5 nm.

Figure 1: (a) Fabrication processes of the Ag nanohemisphere on mirror (NHoM) structure. Top view (b) and side view (c) of the scanning electron microscopy (SEM) images for the Ag nanohemisphere fabricated on the GaN substrate. (d) Top view of the SEM image of the two-dimensional (2D) nanosheet structures of the Ag nanoparticles on silicon substrate reported in [33, 34].

3 Results and discussion

3.1 Optical properties of NHoM

Figure 1(b) shows the SEM image of fabricated Ag nanohemisphere on the GaN substrate which was used instead of BK7 glass to avoid the charge-up effect of the SEM image. Almost the same nanostructures should be formed on BK7 glass substrates. Figure 1(c) shows the side view of the SEM image of the same structure. The nanohemispherical structures were formed with this method. For comparison, the SEM image of the two-dimensional (2D) nanosheet structures of Ag nanoparticles on silicon substrate reported in [33, 34] is also shown in Figure 1(d). The Ag nanoparticles with 5 nm diameter were synthesized by the thermal reduction of a silver acetate precursor. The stable, close-

packed, 2D sheet structures of the Ag nanoparticles were fabricated at an air/water interface via hydrophobic interactions in a Langmuir Blodgett (LB) trough and were transferred onto substrates. Figure 2(a) shows the extinction spectrum converted from the acquired transmission spectrum of the fabricated Ag NHoM structure with a thickness of 10 nm of the SiO₂ spacer layer. The extinction spectrum for the 2D Ag nanosheet structure on quartz substrate [33, 34] is also shown in Figure 2(a). Both spectra contain peaks that are attributed to the localized SP resonance mode at approximately 500 nm. The spectra of the 2D nanosheet were much sharper than that of the nanohemisphere because the shapes and sizes of the Ag particles synthesized by chemical reaction were a) almost spherical with diameters of approximately 5 nm, and b) well-aligned as shown in the SEM image. Compared with the 2D nanosheet structure, the size and shape of the nanohemisphere fabricated by thermal annealing of the metal thin film were random and not aligned, as shown in Figure 1(b). This is why the linewidth of the NHoM peak was much wider than that of the 2D nanosheet structure.

The NHoM spectrum shown in Figure 2 (a) contains the shoulder peak at a shorter wavelength (~350 nm). This peak should be attributed to the quadrupolar oscillation mode generated in the nanohemisphere, while the main peak at ~500 nm was attributed to the dipole oscillation mode. We also measured the extinction spectra of the similar nanohemisphere on sapphire and indium tin oxide (ITO), as shown in Figure 2(b). Basically, the quadrupole mode cannot directly resonate with light because it is a non-radiative mode. However, when the nanohemisphere structures were located on substrates with various refractive indices, the electromagnetic oscillation becomes asymmetric, and the quadrupolar mode can resonate to irradiated light waves. The refractive indices of BK7, sapphire, and ITO at 500 nm, were 1.52, 1.77, and 1.96, respectively. We found that the peak wavelengths of the dipole modes become longer, and the peak intensities of the quadrupolar mode become larger when the refractive index of the substrate is increased. The sharp resonance peaks of the quadrupole modes were superimposed on the broad resonance peak of the dipole mode by Fano resonance and the spectra became asymmetric. For this reason, the peak wavelengths of the quadrupole modes were actually shifted by the refractive index of the substrate, despite the fact that their appearances did not change as shown in Figure 2(b).

We recently found unusual optical properties associated with the multilayers of the Ag nanosheet (3D nanosheet) located on metal substrates with thicknesses of ~100 nm [34]. The peaks of the extinction spectra were split

Figure 2: (a) Extinction spectra acquired from the nanohemisphere on the BK7 glass substrate and the 2D nanosheet structures of the Ag nanoparticles on quartz substrate. (b) Extinction spectra of Ag nanohemisphere on BK7 glass, sapphire, and indium tin oxide (ITO) substrates.

into two other peaks. However, this phenomenon was not observed in the case of the 3D nanosheet on a transparent substrate [34]. This optical phenomenon should be attributed to the mode splitting effect induced by the strong coupling, and suggests that the 3D Ag nanosheet structures can act as plasmonic metamaterials. Given that the size of the Ag nanoparticles in the nanosheet is much smaller than the wavelength of light, it can be approximated as a uniform material by approximating the effective medium. It is thus considered as a type of plasmonic metamaterial. Conversely, the particle sizes of the nanohemispheres are much larger than that of the nanosheet, and they are comparable to the wavelength scale. Therefore, they cannot be approximated as a uniform medium. However, random and large-sized nanohemispheres have similar extinction spectra despite the fact with much broader spectral widths, as shown in Figure 2(a). We thought that

the unique and similar optical properties would be obtained on metal substrates even when nanohemispheres are used which can be fabricated in an easier manner.

Figure 3(a) shows the extinction spectrum converted from the measured reflection spectrum of the fabricated Ag NHoM with a thickness of 10 nm in the spacer layer. The extinction spectrum of NHoG is also shown in Figure 2(a). As shown, the peak of the NHoM is split into two new peaks with higher intensity and increased sharpness, as expected. This suggests that the nanohemispherical structures can also act as plasmonic metamaterials, even if the structures are larger and random. The peak intensity of the long branch peak (at ~600 nm) of the NHoM spectrum was four times higher and its linewidth was halved compared to the peak of NHoM spectrum. This suggests that the SP resonance effect should become eight times higher because the Purcell factor of the cavity is proportional to the Q factors and the inverse of the mode volume. The short branch peak around 380 nm was much stronger and sharper. Wavelengths shorter than 400 nm cannot be easily adjusted with the use of the Ag nanostructures on substrates. The NHoM structure should be useful to enhance the optical properties of UV wavelength regions.

Figure 3(b) shows the calculated extinction spectra by the FDTD simulations for the Ag NHoG and NHoM structures. The diameter of the nanohemisphere was set as 50 nm, and the thickness and the refractive index of the spacer layer were set to 10 nm and 1.5, respectively. The calculated spectra were well matched to the experimental results. The linewidths of the peaks of both branches were much smaller than the experimental results. The extinctions were almost zero between 400 and 550 nm because the calculations were carried out for the single nanohemisphere. Figure 4(a) and (b) respectively show the schematics of the NHoG structure and the spatial distribution of the electric field of horizontal component around the NHoG structure at the peak wavelength (500 nm). The dipole mode of the localized SP mode was clearly visible. Figure 4(c) and (d) respectively show the schematics of the NHoM structure and the spatial distribution of the electric field of horizontal component around the NHoM structure at 500 nm. The dipole mode of the localized SP resonance almost disappeared at this wavelength. Figure 4(e) and (f) show the spatial distribution of the electric field around the NHoM structure at the short (380 nm) and long branches (600 nm), respectively. The dipole mode generated in the nanohemisphere was coupled with the mode generated in the 100 nm Ag layer, and induced quadrupolar-like oscillations. The mode coupling of the short and long

Figure 3: Experimental (a) and calculated (b) results of the extinction spectra of nanohemisphere on glass (NHoG) and the nanohemisphere on mirror (NHoM).

branches seem to be symmetric and antisymmetric mode couplings, respectively. This phenomenon seems similar to the phenomenon at which two atomic orbitals construct bonding and antibonding molecular orbitals. For this reason, we consider that the observed phenomenon is attributed to an electromagnetic mechanism that is analogous to a quantum mechanism. The spatial distribution of the electric field shown in Figure 4 contains small fluctuation especially at the metal interfaces. Such inhomogeneous and singular structures should be numerical artifacts of due to the use of negative dielectric function in the metal and not taken into account hereafter.

We used single hemisphere model with periodic boundary condition as the FDTD simulation, while the fabricated sample has broad inhomogeneity of the size and

Figure 4: (a) Schematic of the nanohemisphere on glass (NHoG) structure. (b) Spatial distribution of the electric field of horizontal component around the NHoG at 500 nm. (c) Schematic of the nanohemisphere on mirror (NHoM) structure. Spatial distribution of the electric field of horizontal component around the NHoM at 500 nm (d), 380 nm (e), and 600 nm (f).

shape of hemispheres. This inhomogeneity might be problem because the SP resonance peak strongly depend on the size and shape. By this reason, the experimentally obtained spectra were broader than the calculated spectra as shown in Figure 3. However, the resonant peaks were still sharp because we could roughly control the size and densities of the hemisphere array structures by controlling the initial thickness of the metal layer and annealing temperatures. Moreover the peaks became much sharper by from NHoM structures because the distances of the nanogaps are exactly same even the sizes and shapes were inhomogeneous. This must be one of the important advantages of this structures.

3.2 Comparison with previously reported NPoM

Despite its very simple structure, the peak splitting of the NHoM extinction spectrum has never been observed and reported. Numerous theoretical and experimental studies have been published with similar metal nanoparticles on mirror (NPoM) structures [40–49]. In many cases, nanosphere on mirror (NSoM) structures were used to obtain very small SP mode nanogaps generated between the metal nanoparticles and metal substrates to obtain strong surface-enhanced Raman spectroscopy (SERS) signals [40–42]. Chikkaraddy et al. used NSoM structures as nanocavities to enhance the emission from point-light emitters, such as single molecules [43]. The metal nanospheres are usually located on the metal substrate. Metal nanospheres that are in contact with the metal substrate at one point should be most suitable for the NSoM structures to obtain the smaller nanogap mode. However, mode splitting, enhancement, and the sharpening of the resonant peak may not occur in the cases of nanospheres located on the metal substrate. Figure 5 shows the shape dependence of the metal nanoparticle on the extinction spectra. The shapes gradually changed from spherical (100%) to hemispherical (50%). In the cases of true spherical shapes, only one peak appeared, and the mode splitting was not observed. The mode splitting effect began to occur, and the splitting energy broadened as the shape approached a hemispherical shape. The electric field in the localized SP mode was strongest near the equator of the sphere. In this way, the mode coupling becomes more prominent in the hemispherical structure where it is closest to the substrate. The metal nanohemisphere is necessary for this phenomenon. This is possibly the reason why this phenomenon has never been observed previously.

Several studies have been reported for NPoM structures with nonspherical nanoparticles. Hung et al. reported a numerical study on three NPoM structures with different shapes: spherical, rods, and cubes on mirror, which have point, line, and plane contacts, respectively [44]. Contrary to intuition, the cube-on-mirror had the smallest mode volume and induced the highest electric field enhancement effect. This result seems consistent with the observed peak splitting owing to the strong mode coupling in the hemisphere on the mirror, but not in the case of the sphere on the mirror. Nanocubes on mirror structures were also used for robust SERS substrates [45] and for nanowaveguides and antennas [46]. The SP resonance peak was observed to be split into two new peaks when dimers were formed, even when nanospheres were used [47]. Plasmon mode

Figure 5: Dependency on the extinction spectra of the various shapes of the nanohemisphere on mirror (NHoM) (spherical (100%) to hemispherical (50%)) obtained with finite difference time domain (FDTD) simulations.

hybridization caused by coupling between the propagation and localized modes of SP have also been reported with nanoplates on mirror structures [48, 49]. These reported hybridizations of the SP modes are based on strong couplings similar to the peak splitting effect observed in this study. However, the peaks were very broad, and the increases of the intensity and sharpness of the peaks were not observed.

Some peak splitting reports have been published for similar structures to that of our NHoM. Huang et al. reported a numerical study of NSoM structures and showed the influence of small morphological changes of NSoM cavities on the near-field enhancement [50]. It was found that the main resonant peak could be tuned linearly when the facet width of the nanosphere increased. The wavelength increased and the peak near-field intensity decreased slightly. This trend seems similar to the trends of the results plotted in Figure 5. However, the peaks were much broader than those observed in this study. The numerical study of the almost similar NHoM structure was also reported by Tserkezis et al. [51]. The peak splitting of the SP mode was obtained, and the two different sets of modes were identified: a) the standard longitudinal gap antenna modes and b) the transverse cavity modes excited in the gap. A similar numerical study was also performed

for the cylindrical nanopatch antennas on mirror structures [52]. The peak splitting was obtained from this structure and was explained in the form of gap modes and antenna modes. These reports published only calculated results, and the expected peak splitting had not been experimentally obtained by the actual fabrication of the NHoM structures. Quite recently, Shi et al. reported peak splitting that was attributed to mode coupling, as experimentally observed with the Au nanoparticles/TiO₂/Au films [53]. In this study, peak splitting should not be expected because the Au nanospheres located on TiO₂ were used. Fortunately, however, half of the Au spheres were buried in TiO and formed hemisphere-like structures similar to our NHoM. This could be the reason why the peak splitting was observed in this structure. The observed split peaks were very broad and their presence was explained by the strong coupling between the Fabry–Pérot nanocavity modes and the localized SP resonant mode. As described above, numerous reports have been published based on calculations and experiments on the peak splitting of the SP mode. However, the enhancement and the increased sharpness of the peaks observed in this study had not been previously observed.

3.3 Flexible tuning of resonance spectrum with NHoM

We have found that the split width of peak splitting owing to mode coupling strongly depends on the distance between the vibration mode of the electric field and the metal substrate. This fact suggests that the SP resonance peak can be tuned by varying the distance between the nanohemisphere and the metal substrate, that is, the thickness of the spacer layer. Usually, the shape and size of the metal nanostructure must be changed to tune the SP resonance mode. If the SP resonant mode can be adjusted only based on the thickness of the spacer layer without changing the shape and size of the metal nanostructure, flexible SP resonance tuning over a large area becomes very easy.

Figure 6(a) shows the results of FDTD calculations of extinction spectra when the spacer thickness of the NHoM structure was changed from 0 to 20 nm. As expected, it was found that the splitting energy of the SP mode strongly depends on the spacer thickness. As the spacer becomes thinner, the two modes come closer to each other, the mode coupling becomes stronger, and the splitting width increases. When the spacer thickness is 5 nm, the splitting width exceeds 300 nm, which is the largest level of mode splitting reported thus far. Figure 6(b) shows the experimental results of the spacer thickness dependence of the

extinction spectra acquired from the NHoM structures. Flexible SP resonance tuning by spacer thickness estimated by calculation was verified by the experimental results. Given that the thickness of the spacer can be easily controlled by a film thickness meter using a quartz oscillator, the SP resonance mode of a large area can be easily tuned with the use of the NHoM structure. Figures 5 and 6 show that the short branch peak around 380 nm were not so changed by the variation of the spacer thickness compared to that of the long branch peak, because the shortest wavelength limit of the SP mode in Ag on glass substrate is around 380 nm. Because of this limitation, the flexible tuning of the SP resonance wavelength in UV regions are still rather difficult compare to the visible wavelength regions by this limit.

Figure 6 shows that not only the peak position of the SP resonance but also the intensity and line width vary strongly as a function of the spacer thickness. This phenomenon cannot be explained solely by the strength of the mode coupling effect because a thinner spacer does not merely cause increased peak sharpness but rather elicits a complex dependence. It is probably necessary to take into account the longitudinal interference of the coupled modes to explain this dependency. The vertical cavity mode should be generated in the nanogap between the metal hemisphere and the metal substrate. This vertical cavity mode is strongly confined into the small mode volume between the nanogap and have a high *Q* value, which depends on the gap distance. The fact that the line width observed in Figure 6 was determined by the spacer

Figure 6: Dependency on the extinction spectra as a function of spacer thickness (from 0 to 20 nm) of the nanohemisphere on mirror (NHoM) structure obtained based on finite difference time domain (FDTD) simulations (a) and experimental measurements (b).

thickness suggests that the vertical nanogap mode should contribute to the resonance spectra. Even if the hemispheres are inhomogeneous in size and shape, the same distance of the nanogaps should be able to give sharp resonance peaks, which is also one of the most important advantages of this structure.

Figure 6 shows that the peak intensity and increased sharpness of the long branch peak could attain their maximum values for spacer thickness values in the range of 10 and 20 nm. Therefore, we calculated the SP resonance spectrum with FDTD simulations by changing the spacer thickness from 13 to 21 nm in steps of 1 nm, as shown in Figure 7. The highest and sharpest peak of the SP resonance was obtained with a spacer thickness of 17 nm. The peak intensity of the extinction reached the value of six, and the Q value estimated based on the linewidth was over 200. These values are surprisingly high for the localized SP mode with metal nanoparticles. These surprisingly high-intensity Q peaks have not been previously obtained experimentally. The NHoM structure generates dark modes by the strong mode coupling through the mirror image and also the vertical nanogap modes strongly confined into the nanogap, and these synergistic should courses the obtained high Q value. To observe this type of ultrasharp spectrum of random NHoM structures, it is necessary to eliminate variations in the size, shape, and arrangement of the metal nanohemisphere. Demonstration of this phenomenon requires that a single nanohemisphere must be excited and measured under a microscope, or that NHoM structures with strictly adjusted size, shape, and alignment must be fabricated by top-down nanofabrication technologies. Given that this structure has a high- Q value and a mode volume smaller than the wavelength, it can be applied to nanolasers.

We expect that the extremely strong interaction characteristics of the NHoM structure will dramatically improve the performance of plasmonics applications, and will overcome the various limitations of optical technology. For example, it is expected that the spatial resolution can be dramatically improved by placing a sample on NHoM and performing a tip-enhanced near-field optical microscope with a metal nanoprobe. If light-emitting materials, such as semiconductor quantum wells and quantum dots can be arranged within the spacer of the NHoM structure, ultrahigh-speed luminescence with a high value **of the Purcell factor** can be expected in addition to the dramatic improvements in luminous efficiency. This feature would realize hyper spontaneous emissions [54], which will provide ultra-fast LEDs with shorter pulses than lasers. Such an incoherent short pulse light source is expected to bring dramatic development in optical communication and

Figure 7: Dependency on the extinction spectra as a function of spacer thickness (from 13 to 21 nm in steps of 1 nm) of the nanohemisphere on mirror (NHoM) structure obtained based on finite difference time domain (FDTD) simulations.

information processing. Furthermore, we expect that the strong coupling regime between excitons and SPs will lead to novel quantum states called plexitons [55], which will enable us to obtain various quantum plasmonic effects. An even more important development refers to the extension of the SP resonance to a wide wavelength range from ultraviolet to infrared. Usually, the wavelength is tuned by controlling the sizes and shapes of the metal species and metal nanostructures, but the tunable width is limited. If the NHoM structure is used, the resonance spectrum can be tuned by varying the thickness of the spacer layer in addition to the metal species. For example, by synthesizing the NHoM structure with aluminum that has SP characteristics in the ultraviolet wavelength range, a very strong and sharp resonance spectrum can be obtained in the deep ultraviolet wavelength range. This will develop the new application fields of deep UV plasmonics, and will lead to the development of high-efficiency deep UV LEDs.

4 Conclusions

In this study, we introduced the metal NHoM structures that have very unique and unusual optical properties. The SP resonant peak was split into two in the UV and visible regions, while only one peak was observed in the case of the Ag NHoG. The split peak wavelengths and intensities were dramatically changed by the thicknesses of the SiO_2 spacer layers. Very strong and sharp peaks were also obtained in the UV region. It has been difficult to obtain these strong and sharp SP resonance spectra in the UV region. These results were reproduced with FDTD simulations. The

calculated results suggest that the long-branch peaks became sharper and stronger, while the Q value reached surprisingly high values (>200) following the strict optimization of the spacer thickness.

Use of nanostructures associated with strong interaction properties, including our proposed NHoM, plasmonics could be able to overcome various limits of conventional optical technologies, such as the spatial, time, and wavelength limits. Given that these properties are related to the performance of all-optical technologies, it has the potential to dramatically advance conventional optical technologies, such as efficient energy devices, threshold-less nanolasers, high-resolution imaging/fabrication, and high-sensitivity sensors. Moreover, it would lead to the development of future technologies, such as highly integrated nano-optical circuits and nano-optical computing. The next-generation optical technologies are expected to become one of the fundamental technologies that will support the emerging ultrasmart and internet-of-things societies. Accordingly, plasmonics is expected to develop further in the future.

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