

## Appeal decision

Appeal No. 2014-8695

Ibaraki, Japan

Appellant

NATIONAL INSTITUTE FOR MATERIALS SCIENCE

The case of appeal against the examiner's decision of refusal of Japanese Patent Application No. 2009- 97564, entitled "Sheet-like Illuminant" (the application published on November 4, 2010, Japanese Unexamined Patent Application Publication No. 2010-248325) has resulted in the following appeal decision.

### Conclusion

The appeal of the case was groundless.

### Reason

#### No. 1 History of the procedures

The application was filed on April 14, 2009, a notice of reasons for refusal was issued on March 11, 2013, and despite the fact that the written opinion and the written amendment were submitted on May 17, 2013, which was within the designated period, a decision for refusal was issued on February 6, 2014, and an appeal against the examiner's decision of refusal was requested on May 12, 2014.

#### No. 2 The Invention

The inventions relating to Claims 1 through 7 of the application are exactly what are specified by the matters described in Claims 1 through 7 of the scope of claims amended by the written amendment dated on May 17, 2013. The invention related to Claim 1 (Hereinafter, simply referred to as "the Invention".) is as follows.

"[Claim 1]

A sheet-like illuminant that emits fluorescence from an entire surface formed by dispersing fluorescent silicon nanoparticles in a transparent resin for maintaining stability of fluorescence and molding the resin into a sheet-like shape."

#### No. 3 Outline of reasons for the examiner's decision

Reasons for the examiner's decision are reasons 1, 2 described in a notification

of reasons for refusal dated on March 11, 2013, namely, the Invention described above is the invention described in a Cited Document 1, which is a Publication distributed in Japan or abroad before the application, namely, National Publication of International Patent Application No. 2006-513458 (hereinafter, referred to as "the Cited Publication"), therefore, since Article 29(1)(iii) of the Patent Act is applicable, the appellant should not be granted a patent, and since the Invention is something that could be easily invented before the application by a person skilled in the art (hereinafter, referred to as "a person skilled in the art") on the basis of the invention described in the Cited Publication, the appellant should not be granted a patent under the provisions of Article 29(2) of the Patent Act.

#### No. 4 Judgment on the body

The body judges that the reasons for refusal of the examiner's decision are still valid even after taking the content of the request for appeal submitted on May 12, 2014 into consideration.

The reasons are as follows.

#### 1 Described matters of the Cited Publication

In the Cited Publication (National Publication of International Patent Application No. 2006-513458, the following matters are described.

##### (1) "[The scope of claims]

###### [Claim 1]

A quantum dot comprising:

- a) a core including a semiconductor material Y selected from the group consisting of Si and Ge; and
- b) a shell surrounding said core, said quantum dot being substantially defect free such that said quantum dot exhibits photoluminescence with a quantum efficiency that is greater than 10 percent.

###### [Claim 2]

The quantum dot of claim 1, wherein Y is Si, and said core has a diameter between approximately 1 nm and 20 nm.

...

###### [Claim 13]

The quantum dot of claim 1, further comprising at least one surface ligand coupled to said shell.

...

[Claim 15]

A quantum dot comprising:

- a) a core including a semiconductor material Y selected from the group consisting of Si and Ge; and
- b) a ligand layer surrounding said core, said ligand layer including a plurality of surface ligands, said quantum dot exhibiting photoluminescence with a quantum efficiency that is greater than 10 percent.

...

[Claim 19]

The quantum dot of claim 15, wherein a surface ligand of said plurality of surface ligands is selected from the group consisting of alkyls, alkenyls, alkynyls, aromatics, aromatic heterocycles, conjugated aromatics, polyenes, cyanides, hydroxys, alkoxys, carboxylates, phenoxys, siloxys, cyanates, thioalkyls, thioaryls, thiocyanates, silylthios, substituted silyl groups, amino groups, mono-substituted amines, di-substituted amines, imino groups, and silylamino.

...

[Claim 22]

A nanocomposite material comprising:

- a) a plurality of quantum dots, said plurality of quantum dots including a semiconductor material Y selected from the group consisting of Si and Ge, at least one quantum dot of said plurality of quantum dots exhibiting photoluminescence with a quantum efficiency that is greater than 10 percent; and
- b) a plurality of molecules coupling said plurality of quantum dots to form one of a linear array, a two-dimensional array, and a three-dimensional array.

...

[Claim 70]

An optical device comprising: a film formed of a nanocomposite material, said nanocomposite material including a plurality of quantum dots, a quantum dot of said plurality of quantum dots including a core that includes a semiconductor material selected from the group consisting of Si and Ge, said quantum dot exhibiting photoluminescence with a quantum efficiency that is greater than 10 percent."

## (2) "FIELD OF THE INVENTION

[0001]

The invention relates generally to quantum dots. More particularly, the

invention relates to quantum dots, nanocomposite materials with quantum dots, optical devices with quantum dots, and related fabrication methods."

(3) "[0017]

Previous attempts at forming quantum dots have largely focused on quantum dots of direct band gap semiconductor materials, such as Group II-VI semiconductor materials. In contrast to such direct band gap semiconductor materials, Group IV semiconductor materials such as Si and Ge have energy gaps, chemical properties, and other properties that render them more desirable for a variety of applications. However, previous attempts at forming quantum dots of Si or Ge have generally suffered from a number of shortcomings. In particular, formation of quantum dots of Si or Ge sometimes involved extreme conditions of temperature and pressure while suffering from low yields and lack of reproducibility. And, quantum dots that were produced were generally incapable of exhibiting adequate levels of photoluminescence that can be tuned over a broad spectral range. Also, previous attempts have generally been unsuccessful, in producing quantum dots of Si or Ge that are sufficiently stable under ambient conditions or that can be made sufficiently soluble in a variety of matrix materials.

[0018]

It is against this background that a need arose to develop the quantum dots, nanocomposite materials, and optical devices described herein."

(4) "[0046]

As used herein, the term "photoluminescence" refers to the emission of light of a first wavelength (or range of wavelengths) by a substance (e.g., a quantum dot) that has been irradiated with light of a second wavelength (or range of wavelengths). The first wavelength (or range of wavelengths) and the second wavelength (or range of wavelengths) can be the same or different.

[0047]

As used herein, the term "quantum efficiency" refers to the ratio of the number of photons emitted by a substance (e.g., a quantum dot) to the number of photons absorbed by the substance."

(5) "(Quantum Dots)

[0078]

Embodiments of the present invention, in part, exploit the extraordinary

properties of quantum dots. Quantum dots have optical and electronic properties that can be dependent (sometimes strongly dependent) on both the size and the material forming the quantum dots.

[0079]

In nature, it is the size range on the order of a few nanometers in which the quantum mechanical characteristics of atoms and molecules often begin to impact and even dominate the classical mechanics of everyday life. In this size range, a material's electronic and optical properties can change and become dependent on size. In addition, as the size of a material gets smaller, and therefore more atomic-like, many characteristics change or are enhanced due to a redistribution of oscillator strength and density of states. These effects are referred to as "quantum confinement" effects. For example, quantum confinement effects can cause the energy gap of the quantum dot or the energy of the light emitted from the quantum dot to increase as the size of the quantum dot decreases. These quantum confinement effects result in the ability to finely tune many properties of quantum dots (e.g., optical and electronic properties) by carefully controlling their size. This control provides one critical aspect of some embodiments of the present invention.

[0080]

A quantum dot will typically be in a size range between about 1 nm and about 1000 nm in diameter or any integer or fraction of an integer therebetween. Preferably, the size will be between about 1 nm and about 100 nm, more preferably between about 1 nm and about 50 nm or between about 1 nm to about 20 nm (such as about 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, or 20 nm or any fraction of an integer therebetween), and more preferably between about 1 nm and 10 nm.

[0081]

Figures 1 (a), 1 (b), 1 (c), and 1 (d) illustrate quantum dots according to some embodiments of the invention. In particular, Figure 1 (a) illustrates a quantum dot 100 comprising a core 102, according to an embodiment of the invention. A core (e.g., the core 102) of a quantum dot may comprise inorganic crystals of Group IV semiconductor materials including but not limited to Si, Ge, and C; Group II-VI semiconductor materials including but not limited to ZnS, ZnSe, ZnTe, ZnO, CdS, CdSe, CdTe, CdO, HgS, HgSe, HgTe, HgO, MgS, MgSe, MgTe, MgO, CaS, CaSe, CaTe, CaO, SrS, SrSe, SrTe, SrO, BaS, BaSe, BaTe, and BaO; Group III-V semiconductor materials including but not limited to AlN, AlP, AlAs, AlSb, GaN, GaP, GaAs, GaSb, InN, InP, InAs, and InSb; Group IV-VI semiconductor materials including but not limited to PbS, PbSe, PbTe, and PbO; mixtures thereof; and tertiary or alloyed compounds of any combination

between or within these groups. Alternatively, or in conjunction, a core can comprise a crystalline organic material (e.g., a crystalline organic semiconductor material) or an inorganic and/or organic material in either polycrystalline or amorphous form.

[0082]

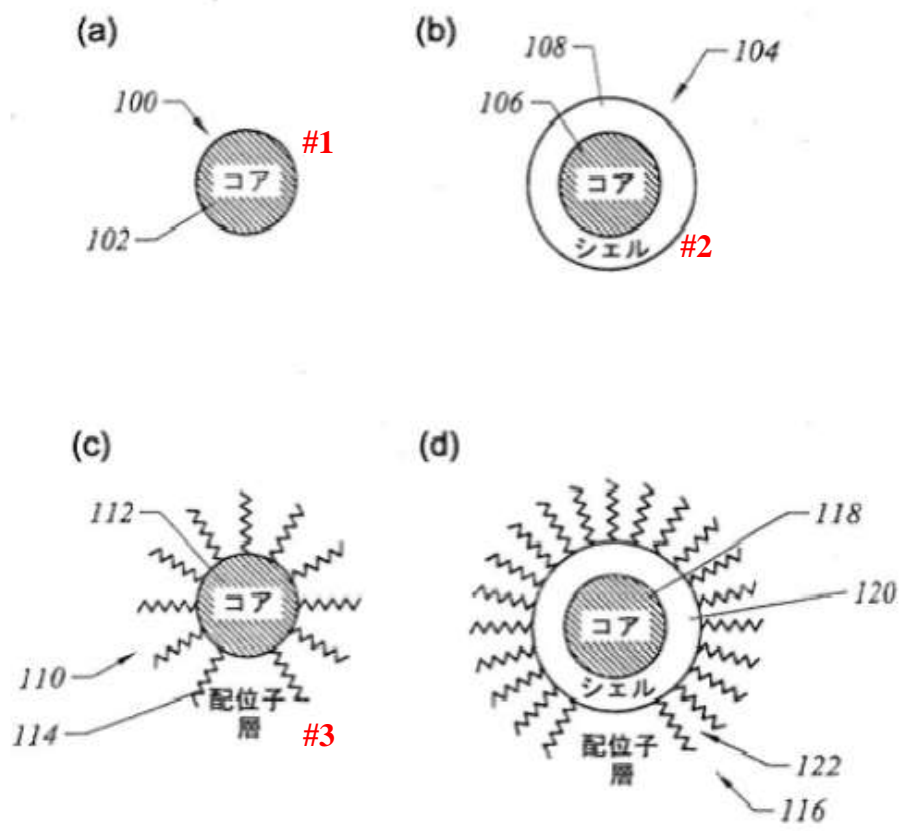
A core may optionally be surrounded by a shell of a second organic or inorganic material. Figure 1 (b) illustrates a quantum dot 104 according to another embodiment of the invention. Here, the quantum dot 104 comprises a core 106 that is surrounded by a shell 108. A shell (e.g., the shell 108) may comprise inorganic crystals of Group IV semiconductor materials including but not limited to Si, Ge, and C; Group II-VI semiconductor materials including but not limited to ZnS, ZnSe, ZnTe, ZnO, CdS, CdSe, CdTe, CdO, HgS, HgSe, HgTe, HgO, MgS, MgSe, MgTe, MgO, CaS, CaSe, CaTe, CaO, SrS, SrSe, SrTe, SrO, BaS, BaSe, BaTe, and BaO; Group III-V semiconductor materials including but not limited to AlN, AlP, AlAs, AlSb, GaN, GaP, GaAs, GaSb, InN, InP, InAs, and InSb; mixtures thereof; and tertiary or alloyed compounds of any combination between or within these groups. Alternatively, or in conjunction, a shell can comprise a crystalline organic material (e.g., a crystalline organic semiconductor material) or an inorganic and/or organic material in either polycrystalline or amorphous form. A shell may be doped or undoped, and in the case of doped shells, the dopants may be either atomic or molecular. A shell may optionally comprise multiple materials, in which different materials are stacked on top of each other to form a multi-layered shell structure.

[0083]

As illustrated in Figures 1 (c) and 1 (d), a quantum dot may optionally comprise a ligand layer comprising one or more surface ligands (e.g., organic molecules) surrounding a core, according to some embodiments of the invention. In Figure 1 (c), a quantum dot 110 comprises a core 112 and a ligand layer 114 surrounding the core 112. In Figure 1 (d), a quantum dot 116 comprises a core 118 and a ligand layer 122 surrounding the core 118. Here, the quantum dot 116 also comprises a shell 120 surrounding the core 118, where the shell 120 is positioned between the core 118 and the ligand layer 122."

[FIG. 1] cited by the summarized matters is as follows.

"[FIG. 1]



- #1 Core
- #2 Shell
- #3 Ligand layer"

(6) "(Optical Properties)  
 (Linear Optical Properties)  
 [0084]

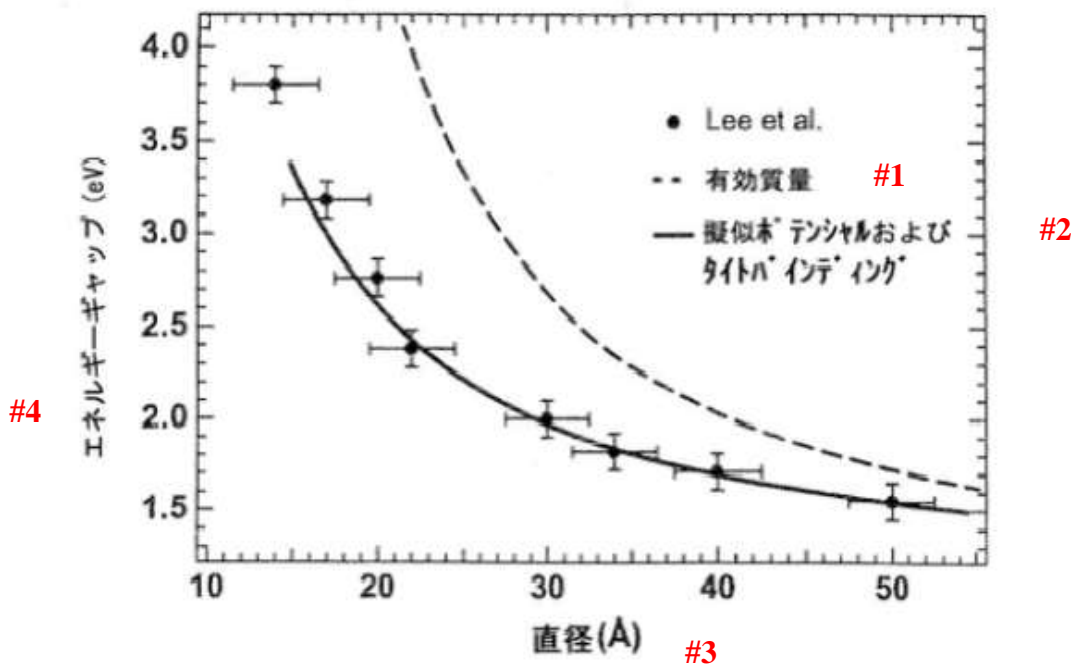
One of the most dramatic examples of "quantum confinement" effects is that, for a semiconductor material, the energy gap shifts as a function of size. This can be seen in Figure 2, where the energy gap of quantum dots fabricated from silicon, referred to herein as "silicon quantum dots", is plotted as a function of the size (e.g., diameter) of the quantum dots, according to an embodiment of the invention. The silicon quantum dots were made as described herein. The vertical axis represents the energy gap of the silicon quantum dots, and the horizontal axis represents the size of the silicon quantum dots. The observed values for the energy gap (dots with error bars) are compared against pseudopotential and tight-binding models (solid line) and against the simple effective mass theory (dashed line).

[0085]

The same effect can be seen for the emission wavelength as a function of the size of quantum dots. Figure 3 illustrates photoluminescence (PL) spectra from six samples with different sizes of silicon quantum dots, according to an embodiment of the invention. The silicon quantum dots were made as described herein and include shells formed of an oxide. The vertical axis represents a normalized PL signal, and the horizontal axis represents the emission wavelength. The PL spectra illustrated in Figure 3 is obtained by optically exciting the silicon quantum dots with ultraviolet light. The wavelength of the optical excitation is shorter than the wavelength at the absorption edge of the silicon quantum dots. Figure 3 demonstrates the range of sizes that can be made with the methods described herein. The quantum dots shown at the top of Figure 3 are not drawn to scale and are meant to illustrate the relative size of the quantum dots responsible for the PL spectra. Figures 2 and 3 demonstrate the unprecedented control that can be obtained over absorption and emission characteristics of the silicon quantum dots."

[FIG. 2] and [FIG. 3] cited by the summarized matters are as follows.

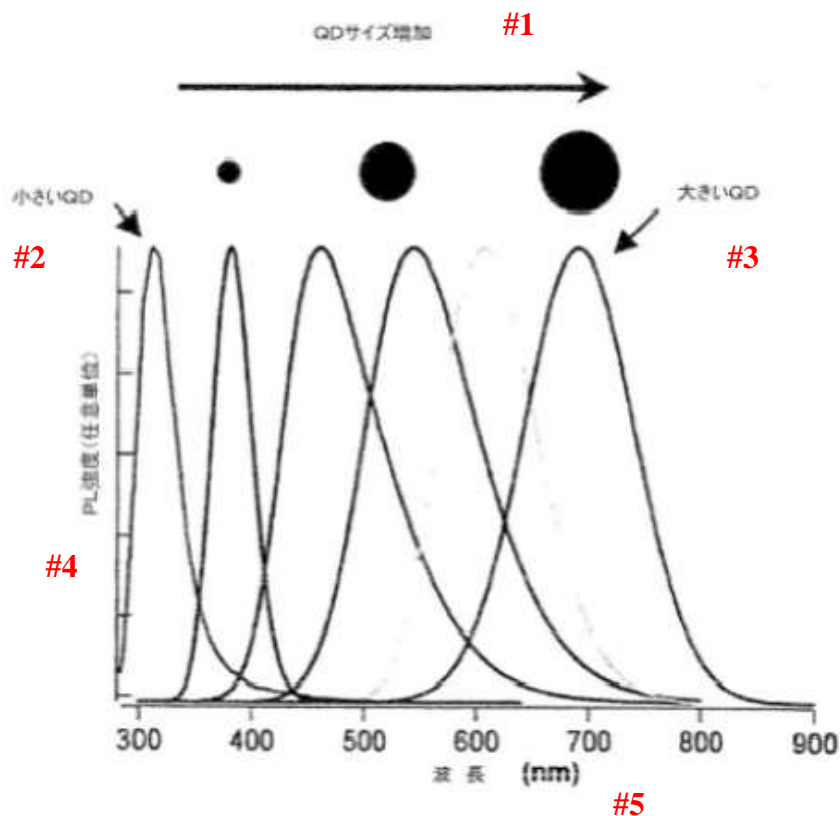
"[FIG. 2]



- #1 Effective mass
- #2 Pseudopotential and Tight binding
- #3 Diameter
- #4 Energy gap

[FIG. 3]





- #1 QD size increase
- #2 Small QD
- #3 Large QD
- #4 PL intensity (arbitrary unit)
- #5 Wavelength"

(7) "(Chemical properties)  
 (Chemically controllable surface)  
 [0117]

According to some embodiments of the invention, a unique physical characteristic of quantum dots is that, while the core can comprise a crystalline semiconductor material, the surface can be coated with a variety of different organic and/or inorganic materials. These surface coatings (e.g., shells or ligand layers) can impart stability and chemical activity, as well as passivation of electrically and optically active defect sites on the quantum dot surface. These surface coatings are optionally substantially different in chemical nature than the inorganic core. As a result, while quantum dots can comprise primarily a highly nonlinear semiconductor material, they substantially appear to the surrounding material as surface ligands. As such, the processability and chemical stability of this highly nonlinear and tunable optical

material can primarily be a function of the surface layer and not a function of the material that provides the majority of the optical characteristics.

[0118]

Surface ligands are preferably bi-functional. By bi-functional, it is meant that there are at least two portions of the surface ligand such that one portion interacts primarily with the quantum dot surface, while the second portion interacts primarily with the surrounding environment (e.g., solvent and/or matrix material). These at least two portions of the surface ligand may be the same or different, contiguous or noncontiguous, and are optionally contained within two or more different molecular species that interact with each other to form the ligand layer. The at least two portions can be selected from a group consisting of hydrophilic groups, hydrophobic groups, or amphiphilic groups. The interaction of each of the at least two portions and the quantum dot or surrounding environment can be covalent or noncovalent, strongly interacting or weakly interacting, and can be labile or non-labile. The at least two portions can be selected independently or together.

[0119]

In some embodiments of the present invention, the surface ligands are selected such that the portion that interacts with the quantum dot passivates defects on the surface such that the surface is made substantially defect-free. At the same time, the portion that interacts with the environment is selected specifically to impart stability and compatibility (e.g., chemical compatibility or affinity) of the quantum dot within a matrix material that is selected for a specific application. Simultaneously satisfying both of these requirements is an important aspect of certain embodiments of the present invention relating to the development of an engineered nonlinear nanocomposite material. Alternative methods of achieving these requirements include, for example, 1) Passivating the surface of the quantum dot independent of the ligand layer (e.g., using a shell or creating an intrinsically defect free surface), while the environmental compatibility is imparted by the surface ligands, or 2) imparting both passivation and environmental compatibility independent of the ligand layer. Achieving passivation of the surface of quantum dots is one advantage of using colloidal quantum dots over alternate approaches.

[0120]

Through the appropriate selection of surface ligands, quantum dots can be incorporated into a variety of matrix materials such as, for example, liquids, glasses, polymers, crystalline solids, and even close-packed ordered or disordered quantum dot arrays. The resulting nanocomposite materials can be formed into homogeneous,

high-quality optical films of quantum dots. Alternatively, the chemistry can be selected to allow dispersion of the quantum dots into a matrix material with a controllable degree of aggregation, forming micron or sub-micron sized clusters. The result is an increased local fill-factor and an enhanced local electric field effect that may further increase the nonlinear response of the nanocomposite materials of embodiments of the present invention."

(8) "(Novel Quantum Dot Materials)

[0172]

In one embodiment, the quantum dots are silicon quantum dots or germanium quantum dots that are surface passivated (or terminated) with an inorganic layer (such as oxides of silicon and germanium) and/or organic and/or inorganic surface ligands, herein sometimes referred to as SiQDs and GeQDs, respectively. SiQDs and GeQDs as described herein are novel types of quantum dots that show definitive quantum confinement effects as manifested by size dependent properties such as size-dependent energy gaps that can be tuned over a very broad range and in particular from the near infrared to the near ultraviolet. In addition, SiQDs and GeQDs are stable under a variety of environmental conditions including ambient (e.g., pressure :-1 atmosphere; Gases :-70% nitrogen,-30% oxygen; Temperature :-20-25 C) for desired periods of time depending on the specific application. A SiQD and a GeQD can be comprised of a substantially Si core for a SiQD and a substantially Ge core for a GeQD. In addition, the "surface" of the SiQD can be comprised of Si and inorganic elements such as oxygen and/or organic ligands (R). In addition, the "surface" of the GeQD can be comprised of Ge and inorganic elements such as oxygen and/or organic ligands (R).

[0173]

In one embodiment of the invention, a SiQD comprises a substantially defect free silicon crystal core of diameter between approximately 1 nm and 100 nm, preferably between approximately 1 nm and 20 nm, more preferably between approximately 1 nm and 10 nm, while a GeQD comprises a substantially defect free germanium crystal core of diameter between approximately 1 nm and 100 nm, preferably between approximately 1 nm and 50 nm, more preferably between approximately 1 nm and 20 nm. In the case of an inorganic shell surrounding the silicon or germanium core, this shell typically has a thickness of between approximately 0.1 and 5 nm. One preferred inorganic shell is SiO<sub>n</sub> for SiQD and GeO<sub>n</sub> for GeQD with n ranging between approximately 0 and 2, preferably ranging between approximately 1.5 and 2, most preferably ranging between approximately 1.8 to 2.

The chemical composition of the shell (e.g., relative amounts of Si (or Ge) and O) is potentially varying continuously through a portion of the shell and optionally varying discontinuously through a portion of the shell, in which case  $n$  can represent an averaged value within the shell. In the case of organic surface ligands terminating the surface, the SiQD and GeQD can comprise ligand layers comprising organic molecules with a structure R. R can be any one of a variety of hydrophobic, hydrophilic, or amphiphilic molecules (a list of preferred surface ligands is included below). The surface ligands can provide a surface coverage of available silicon (or germanium) and oxygen binding sites at the surface to provide between approximately 0% and 100% surface coverage, preferably between approximately 20% and 100% surface coverage, more preferably between approximately 50% and 100% surface coverage, more preferably between approximately 80% and 100% surface coverage, with a maximum of one or more complete layers of surface ligands. R can optionally comprise a plurality of different organic molecules at a plurality of absolute and relative densities. Finally, a SiQD or GeQD may optionally comprise additional R-groups that do not interact directly with the quantum dot surface, but rather indirectly through other R-groups interacting directly with the surface. In this case, surface coverage greater than 100% is possible.

[0174]

It has long been considered that the production of an ambient-stable silicon quantum dot or germanium quantum dot with a defined oxide shell could not be achieved due to difficulties in growing a stable and trap-free surface oxide shell. Thus, the SiQD and GeQD described herein represent a substantial advance."

(9) "(Preferred Surface Ligands and Molecular Tethers)

[0236]

As described in the previous sections, the ligand layer can serve to passivate the surface of a quantum dot and eliminate surface defects. It also facilitates compatibility with matrix materials. This is further explained as follows. Fluoropolymers are a group of desirable materials for optical applications because of their unique properties. Fluoropolymers, in general, have low indices of refraction (e.g., in comparison with regular hydrocarbon polymers) and thus low intrinsic scattering loss. They also, in general, exhibit low absorption loss as they are typically comprised of little or no carbon-hydrogen bonds. They are hydrophobic and thus low in moisture absorption. They, in general, are chemically and thermally inert and thus compatible in demanding environments and extreme process conditions in device fabrication. Because of their

inertness, fluoropolymers are nearly non-mixable with many materials, such as conventional quantum dots. Embodiments of the present invention provide a novel approach to circumvent the compatibility issue by introducing fluorinated surface ligands to the surface of quantum dots (e.g., as in Example 11 and 12 of the preceding section). The quantum dots, which are terminated with a ligand layer of fluorinated surface ligands, can now be incorporated into, for example, Cytop brand polymer (a perfluorinated polymer from Asahi), facilitated, for example, by using a solvent vehicle, CT-SOLV 180 from Asahi.

[0237]

The following are preferred surface ligands of the ligand layer, according to some embodiments of the invention. This list, which is not intended to be exhaustive, describes a number of surface ligands having desirable physical characteristics that can be used to form ligand layers for SiQDs or GeQDs. In the following, Y is Si or Ge, and T-C, Y-O, Y-S, Y-Si, and Y-N denote covalent bonds between Si or Ge and a C atom, an O atom, an S atom, a Si atom, and an N atom, respectively. Other preferred surface ligands, not listed below, can contain a P or a Se atom that is covalently bonded to Si or Ge.

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[0238]

Figures 10 (a) and 10 (b) show PL spectra of organic-terminated Si quantum dots, and Figures 11 (a) and 11 (b) show PL spectra of organic-terminated Ge quantum dots. The Si and Ge quantum dots were made with the methods described herein. The PL spectra show that the light emission can be readily tuned from the red to the ultraviolet by exciting quantum dots of different sizes. The PL spectra are obtained by optically exciting the quantum dots with wavelengths shorter than the wavelength at the absorption edge of the quantum dots. This light emission is stable in ambient conditions. This stability is due in large part to the relative completeness and stability of the surface termination, e.g., the surface termination and the interface between the core and the surface termination is substantially defect free. In Figures 10 (a) and 10 (b), the surfaces of the Si quantum dots are terminated with 4-methoxyphenyl groups. In Figure 10 (b), the vertical axis represents a normalized photoluminescence signal from Figure 10 (a).

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[0239]

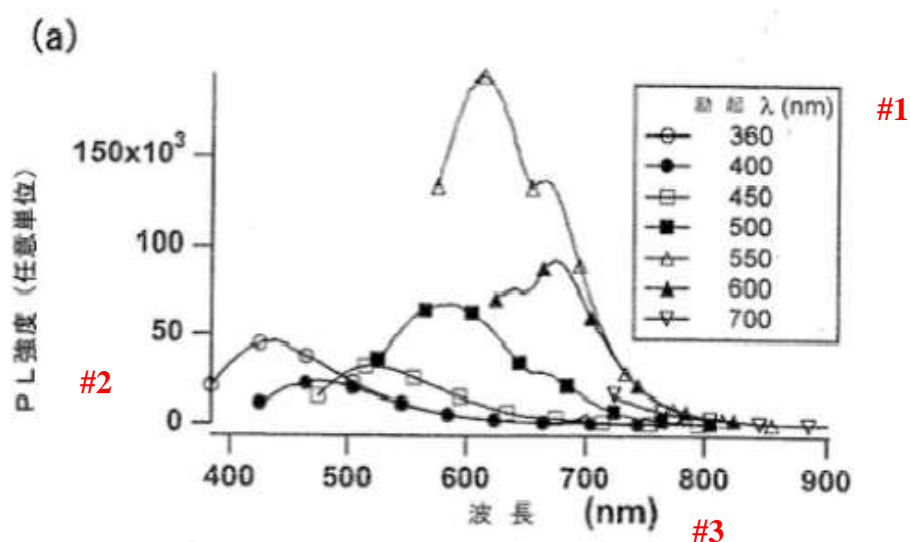
The electronic and optical properties of these organic-terminated SiQDs and GeQDs that are made in this fashion are unique in that they show size dependent

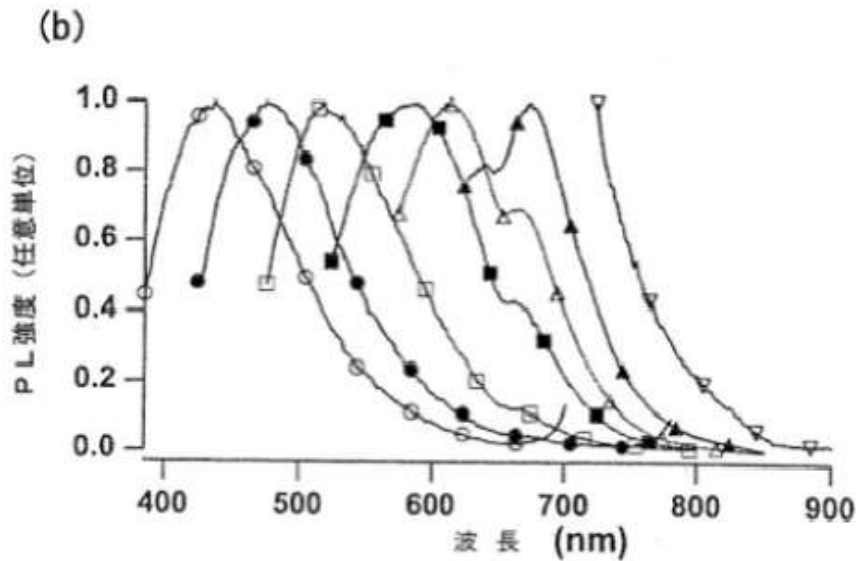
properties that are uniquely consistent with quantum confinement. The optical and electronic properties of these SiQDs and GeQDs are uniquely consistent with theoretical calculations more sophisticated than Effective Mass approaches, such as the Empirical Pseudopotential Method and the Tight Binding Method. A comparison of the size dependent energy gap calculated by these methods with measurements taken on the SiQDs and GeQDs synthesized by the method disclosed herein show that the agreement is extremely good and is the best observed for any quantum dot formed of Si or Ge. [0240]

According to some embodiments of the invention, nanocomposite materials comprising quantum dots that are surface-terminated with various organic groups and dispersed in processable matrix materials such as organic polymers or sol-gels can exhibit new quantum phenomena. These new quantum phenomena in turn allow a large variety of new applications (such as all-optical switching) and the fabrication of device structures using low cost processing techniques (e.g., spin coating or dipping). Described herein are several novel synthetic schemes to fabricate these quantum dots and to functionalize their surfaces with molecular species that are chemically bonded to the surface for stability and robustness. Use of such functionalized quantum dots avoids the need for expensive and specialized fabrication equipment and facilities. The synthesis of these nanostructures can be readily implemented in many laboratories."

[FIG. 10] cited in the summarized matters are as follows.

"[FIG. 10]





- #1 Excitation  $\lambda$
- #2 PL intensity (arbitrary unit)
- #3 Wavelength"

(10) "(Preferred Matrix Materials)

[0245]

According to some embodiments of the present invention, the matrix material that is used to host quantum dots can be selected from a broad range of materials due in large part to the versatile surface termination of the quantum dots as discussed above. These matrix materials can include, for example, organic and inorganic polymers or glasses with different properties including mechanical strength, optical transparency, lightwave transmissivity, thermal stability, dimensional stability, low temperature flexibility, moisture absorption, and chemical inertness.

[0246]

The matrix materials in some embodiments of the present invention are preferred to be highly transparent and low absorption in the wavelength range from 600 nm to 2  $\mu\text{m}$ . Also, they are preferred to be highly compatible with quantum dots so that a desired amount of quantum dots can be readily incorporated into the resulting nanocomposite material without degrading optical and mechanical properties. Polymers with special functional groups may be selected to facilitate solubility interactions and enhance compatibility with quantum dots. This is further explained as follows. Polymers with Lewis acid (base) functional groups, for example, can be selected to host quantum dots which are surface-functionalized with Lewis base (acid)

surface ligands. Another example is to take advantage of hydrogen-bonding interactions. Polymers of hydrogen-bonding donors (acceptors) are the preferred matrix materials for quantum dots that are surface functionalized with hydrogen-bonding acceptors (donors). Additionally, polymers with strong dipolar groups are the preferred matrix materials for quantum dots that are engineered with strong dipolar surface ligands. The strong intermolecular interactions described above greatly enhance compatibility between the quantum dots and the matrix materials. Therefore, high contents of quantum dots can be readily incorporated into the matrix materials while maintaining desired uniformity and homogeneity (e.g., allowing the quantum dots to be substantially uniformly dispersed throughout the matrix materials). Additionally, block copolymers can be used to further enhance compatibility by skillful selection of monomer units and block length. As a result, a nanocomposite material comprising quantum dots and a matrix material can be engineered to be of high optical quality and low scattering loss. More importantly, the linear and nonlinear index of refraction can be tuned for a variety of applications primarily by adjusting the content of the quantum dots and by selecting the matrix material, according to some embodiments of the invention.

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[0254]

Another preferred matrix material is a homopolymer including, but not limited to, the following:

...

- o. Cytop (Cytop) (registered trademark)
- p. PMMA
- q. Fluorinated acrylates
- r. Poly (siloxanes)"

(11) "(Preferred nanocomposite materials)

[0263]

Embodiments of the present invention comprise a nanocomposite material with a controllable set of optical, mechanical, chemical, and electronic properties. The nanocomposite material can comprise quantum dots dispersed in an organic and/or inorganic matrix material.

...

[0264]

The nanocomposite materials according to some embodiments are preferably



optically pure, with a homogeneous distribution of quantum dots dispersed therewithin. These quantum dots may be substantially uniformly dispersed as individual dots or as aggregates of controlled sizes (e.g., smaller aggregates up to massively interconnected quantum dot subsystems). The engineered nanocomposite materials are preferably optically homogeneous and uniform, so that little or no scattering and/or mode disruption result from light passing through or past the material, as the specific application demand. In certain applications, close-packed micron or sub-micron-sized clusters of quantum dots dispersed in a polymer or other matrix material with a filling fraction optimized to enhance local field effects may be preferable.

[0265]

The nanocomposite materials according to some embodiments can exhibit increased photoluminescence quantum efficiency. For certain applications, the nanocomposite materials can comprise quantum dots that are substantially defect free and that collectively exhibit photoluminescence with a quantum efficiency that is greater than 6 percent, preferably greater than 10 percent, more preferably at least 20 percent, more preferably at least 30 percent, more preferably at least 40 percent, and more preferably at least 50 percent.

...

[0270]

In addition, the nanocomposite materials described herein are relatively simple and inexpensive to make, are more easily processed, are compatible with a large number of other material systems, and can be incorporated more readily into various device structure and in nearly any device size. Epitaxial growth of the nanocomposite materials described herein is typically not required, which can be an advantage since epitaxial growth is typically an expensive process that is often not amenable to simple processing or large area devices and is often incompatible with many other material systems (since it requires epitaxial growth on material systems that are lattice-matched to itself). The nanocomposite materials can be deposited on various substrates, substantially independently of their size, surface area, and surface nature, in the form of films or coatings of varying thicknesses and can be formed into structures of various shapes and sizes. Importantly, these films, coatings, and structures made from the nanocomposite materials can be manufactured with a number of simple and inexpensive fabrication techniques such as spin coating, spray coating, doctor blading, and dip coating at ambient temperature and pressure, or using conventional molding processes for engineering plastics and elastomers."

(12) "(Substitute applications of engineered nanocomposite material)  
[0272]

While the application of engineered nanocomposite materials as a nonlinear material is described herein, it should be recognized that such materials will also find applications in a variety of areas such as, though not limited to, engineered resonant nonlinear nanocomposite materials, engineered linear nanocomposite materials, engineered absorptive nanocomposite materials, engineered electro-optic nanocomposite materials, engineered thermo-optic nanocomposite materials, engineered thermal nanocomposite materials, engineered gain nanocomposite materials, engineered magneto-optic nanocomposite materials, engineered magnetic nanocomposite materials, engineered electronic nanocomposite materials, engineered biological nanocomposite materials, engineered optoelectronic nanocomposite materials, and engineered mechanical nanocomposite materials. The tunable physical, chemical, electronic, and optical properties of the materials described herein, as well as the methods of incorporation thereof, can be used to create materials with specific characteristics tailored to many other applications."

(13) "(Integrated optical pump for quantum dot lasers and amplifiers)  
[0396]

Conventional quantum dot based lasers and amplifiers typically suffer from a lack of an efficient means of electrical excitation. One means around this problem is to find an efficient and inexpensive method of optically exciting quantum dots for use as lasers and amplifiers. A novel type of SOA comprises a pump laser (e.g., a 980 nm pump lasers for EDFAs), a device configured to focus a pump beam, and a nanocomposite material according to some embodiments of the invention. This configuration could potentially find application wherever SOAs are currently being considered (e.g., inexpensive multi-wavelength amplifiers, all-optical switches, and wavelength converters, etc.). Additionally, an optically pumped quantum dot laser would require a pulsed laser source (to overcome fast Auger recombination) and a device for producing a laser cavity."

2 The invention described in the Cited Publication (the Cited Invention)

(1) In the Cited Publication, quantum dots and nanocomposite materials having the quantum dots are described (summarized matters (2)), and the quantum dots have shells or ligand layers formed on surfaces thereof (summarized matters (1), (5), (7)) in consideration of the conventional problems, namely, stability under the ambient

conditions or solubility to matrix materials (the summarized matters (3)).

(2) Particularly, that surface ligands constituting the ligand layers passivate surface defects of quantum dots and promote stability and compatibility of the quantum dots in matrix materials is described ([0119] of the summarized matters (7) and [0236] of the summarized matters (9)), organic-terminated silicon quantum dots (Si quantum dot, SiQD) whose surfaces are terminated with organic surface ligands are exemplified as a specific example ([0172] and [0173] of the summarized matters (8) and [0238] of the summarized matters (9)), and that the organic-terminated silicon quantum dots (Si quantum dots, SiQD) exhibit stable photoluminescence under the ambient conditions is described ([0238] of the summarized matters (9) and [FIG. 10]).

(3) Also, nanocomposite materials including quantum dots whose surfaces are terminated with various organic groups, dispersed in matrix materials such as polymers compatible with processing such as spin coating or dipping are described ([0240] of the summarized matters (9) and [0246] of the summarized matters (10)).

(4) Therefore, it can be said that the following invention is described in the Cited Publication (hereinafter, referred to as "the Cited Invention").

"A nanocomposite material formed by dispersing organic-terminated silicon quantum dots exhibiting photoluminescence in a matrix material that is processable such as a polymer."

### 3 Comparison

(1) "The silicon quantum dots" of the Cited Invention are nanometer-order particles ([Claim 2] of the summarized matters (1), [0080] of the summarized matters (5), and [0173] of the summarized matters (8)), and exhibit photoluminescence, namely, fluorescent properties (as for a definition of photoluminescence, refer to the summarized matters (4))

"The fluorescence silicon nanoparticles" of the Invention do not particularly exclude ones whose surfaces are modified with ligand layers and such.

Therefore, it can be said that "the organic-terminated silicon quantum dots exhibiting photoluminescence" of the Cited Invention corresponds to "the fluorescent silicon nanoparticles" of the Invention.

(2) Since "the transparent resin" of the Invention in which fluorescence silicon

nanoparticles are dispersed is generally so called a matrix material, "the transparent resins" of the Invention and "the processable matrix materials such as polymers" have something in common in terms of being matrix materials.

(3) Therefore, it can be said that the Invention and the Cited Invention coincide in terms of "an article formed by dispersing fluorescence silicon nanoparticles in a matrix material" and differ in terms of the following points.

(i) A different feature 1: While the matrix materials of the Invention are transparent resins and maintain the stability of fluorescence of the fluorescence silicon nanoparticles, although the matrix materials of the Cited Invention permit polymers, whether they are transparent resins and whether they maintain stability of fluorescence of the fluorescence silicon nanoparticles are not clear.

(ii) A different feature 2: While the article of the Invention is formed by molding into a sheet-like shape and is a sheet-like illuminant for emitting fluorescence from the entire surface, whether the article of the Cited Invention is formed into a sheet-like shape and whether it is a sheet-like illuminant emitting fluorescence from the entire surface are not clear.

#### 4 Examination of the different features

##### (1) On the different feature 1

A It can be said that, in the summarized matters (10) of the Cited Publication, (i) through (v) below are described concerning the matrix materials related to the Cited Invention.

(i) For matrix materials used for accepting quantum dots, for instance, polymers having properties such as optical transparency or chemical inertness may be used.

(ii) The matrix materials are preferably highly transparent and have low absorption in a wavelength range from 600 nm to 2  $\mu\text{m}$ , and have high compatibility with quantum dots so as not to degrade optical and mechanical properties and so as to be easily integrated into resultantly obtained nanocomposite materials.

(iii) Polymers having special functional groups promote the interactions of solubility and reinforce the compatibility with quantum dots. For instance, polymers having Lewis acid (base) functional groups reinforce compatibility with quantum dots to the surfaces of which a function is given with surface ligands of Lewis acid (base).

(iv) Polymers having strong dipolar groups are preferable matrix materials for quantum dots using strong dipolar surface ligands. Since strong intermolecular interactions significantly reinforce the compatibility between quantum dots and matrix

materials, even quantum dots of high content are easily integrated into matrix materials (for instance, quantum dots are substantially uniformly dispersed in the entire matrix materials) while maintaining desired uniformity and homogeneity.

(v) As preferable matrix materials, transparent resins such as Cytop polymers or Poly (siloxanes) may be used.

B As described above, it can be said that for matrix materials of the Cited Invention transparent resins may be used as polymers (described above in 4(1)(i), (ii), and (v)), and optical transparency is taken into account. Since the quantum dots of the Cited Invention were developed to solve the problems of improving the stability under the ambient conditions and the solubility (compatibility) to matrix materials (summarized matters (3)), it should be said that, needless to say, for matrix materials used in combination with the quantum dots, the problems (particularly, stability) must be considered, the matrix materials are to be so selected as not to degrade optical properties and such of the quantum dots (described above in 4(1)(ii)), and the matrix materials are to be selected by considering the chemical inertness (chemical stability) (described above in 4(1)(i)) and further compatibility (described above in 4(1)(iii) and (iv))) in addition to the optical transparency. Moreover, since, by considering that the matrix materials enclose the dispersed quantum dots (isolate from the ambient conditions), it is reasonably understood that the matrix materials play a role of protecting the quantum dots, and the matrix materials are to be selected by considering also the stability of fluorescence of the quantum dots.

C Therefore, although it is not clearly described whether "the polymers that are matrix materials" of the Cited Invention are transparent and whether they maintain the stability of fluorescence of nanoparticles, since, as described above, such properties are to be naturally considered for the polymers of the Cited Invention, it cannot be said that the different features concerning the properties of the matrix materials are substantial. Even when the different features are assumed to be substantial, with reference to the descriptions above of the Cited Publication, there seem no difficulties in adopting matrix materials having properties described above for the polymers of the Cited Invention. In this case, as described above, since it is understood that the matrix materials protect the peripheries of the quantum dots and contribute to maintain the stability of the fluorescence, it can be said that technical matters of the Invention associated with the different features may be easily conceived by a person skilled in the art.

(2) On different feature 2

A The Cited Invention does not clearly describe the form, whether it is of a sheet-like shape.

However, Claim 70 of the summarized matters (1) and the paragraph [0120] of the summarized matters (7) of the Cited Publication describe that the nanocomposite material is made to be of a film form. Since the paragraph [0270] of the summarized matters (11) of the Cited Publication has a description, regarding the production of nanocomposite material, reading: "the nanocomposite materials can be deposited on various substrates, substantially independently of their size, surface area, and surface nature, in the form of films or coatings of varying thicknesses and can be formed into structures of various shapes and sizes. Importantly, these films, coatings, and structures made from the nanocomposite materials can be manufactured with a number of simple and inexpensive fabrication techniques such as spin coating, spray coating, doctor blading, and dip coating at ambient temperature and pressure, or using conventional molding processes for engineering plastics and elastomers", it can be understood that forms of the nanocomposite materials related to the Cited Invention conceivably include a film-like shape (which can be considered as one of "sheet-like shapes"), and, since, as manufacturing methods, the conventional molding processes of engineering plastics and elastomers in addition to spin coating under ambient temperature and pressure are mentioned, it can be said that manufacturing the film-like (sheet-like) shapes with typical molding processes is within expectation. Even when the Cited Invention does not assume a sheet-like final form as described above, since determining in what shape a final form is to be made is nothing but a matter a person skilled in the art should design according to usages to be used, it can be said that making a final form to be of a sheet-like shape is easily conceivable by a person skilled in the art (refer to the following Reference, if necessary).

- Reference 1: National Publication of International Patent Application No. 2007-523221 (a Cited Document 3 cited by the examiner's decision) (refer to [Claim 15], and [Example 3] to [Example 5])
- Reference 2: Japanese Unexamined Patent Application Publication No. 1997-111232 (refer to the paragraphs [0034], [0044], and [0045], and luminous layers 34, 40 in [FIG. 2] and [FIG. 3])
- Reference 3: International Publication No. WO 2009/002512 (refer to paragraphs [0080], [0086], [0105], [0116], and [0183], and [FIG. 3], [FIG. 5], and [FIG. 6] of one of the patent family, National Publication of International Patent Application No. 2010-532910)

B Since silicon quantum dots in the nanocomposite materials related to the Cited

Invention exhibit stable photoluminescence under the ambient conditions by terminating with organic groups (refer to [0238] and [FIG. 10] of the summarized matters (9)), the matrix materials accepting the same is also selected by considering so as not to degrade optical properties and such of the quantum dots as described above in 4(1)A, it is reasonable to think that, even after being molded into sheet-like final forms by the molding process described above, the emission characteristics are naturally maintained. In such a case, it is reasonable to understand that, by irradiating excitation light of a specific wavelength, silicon quantum dots dispersed in the nanocomposite materials of the Cited Invention exhibit the photoluminescence (predetermined emission characteristics).

C Certainly, the Cited Publication has a description about applications of the nanocomposite materials to optical devices (optical switches, etc.) using nonlinear optical properties (particularly, nonlinear index of refraction) of quantum dots, although not having an explicit description that the nanocomposite materials themselves function as illuminants, [0085] and [FIG. 3] of the summarized matters (6) have a description that luminous wavelengths can be seen as a function of the size of quantum dots (that luminous properties can be controlled by changing the size of quantum dots) as linear optical properties of quantum dots, [0265] of the summarized matters (11) has a description that the nanocomposite materials related to some embodiments exhibit increased photoluminescence quantum efficiencies, the summarized matters (12) has a description reading: "although applications as nonlinear materials for engineered nanocomposite materials are described here, such materials may also have applications in various fields including ●●● engineered linear nanocomposite materials, ●●● engineered electro-optic nanocomposite materials, engineered thermos-optic nanocomposite materials, engineered magneto-optic nanocomposite materials ●●● ", further, the summarized matters (13) have an exemplification on an optically excited quantum dot laser. Therefore, it can be said that the nanocomposite materials related to the Cited Invention are not limited to application examples of the quantum dots exhibiting no fluorescence, but conceivably include application examples of the quantum dots exhibiting fluorescence as well. In addition, as is found in the Reference described above, since illuminants of sheet-like shapes are well known, no particular inventiveness is recognized in utilizing the nanocomposite materials related to the Cited Invention for sheet-like illuminants.

### (3) Summary of the examinations of the different features

As described above, it should be said that it is already conceived that the

nanocomposite materials of the Cited Invention is molded into sheet-like shapes as final forms, even if it is not the case, whether to make them into the sheet-like shapes is simply a matter of designing. It can be also said that it is already conceived that the nanocomposite materials related to the Cited Invention molded into sheet-like shapes in such a way are used as illuminants since the silicon quantum dots themselves dispersed therein exhibit photoluminescence (emission characteristics). Even if it is not conceived that the nanocomposite materials related to the Cited Invention molded into sheet-like shapes is used as illuminants, considering that sheet-like illuminants themselves are already well known, no particular inventiveness can be recognized in using as such illuminants.

## 5 Arguments of appellant

(1) The appellant argues the following points in the request for appeal.

(i) In the Cited Invention, although that quantum dots have fluorescent properties is disclosed, there is no disclosure as to whether, by dispersing the quantum dots into matrix materials, produced nanocomposite material films emit light, and the optical properties of nanocomposite materials used when such kinds of films are applied to optical devices are not the luminous properties but the nonlinearity of index of refraction.

(ii) Since the fluorescent properties of fluorescence silicon nanoparticles is easily lost by oxidation and such, as in the case of the Cited Invention, when quantum dots are applied in a form of a nanocomposite material film and when fluorescent properties themselves of the film are not used, even when the quantum dots, starting materials, have fluorescent properties, in manufacturing the nanocomposite materials and molding into the film, it cannot be expected that attention is paid for maintaining the fluorescent properties, and even when quantum dots having silicon cores disclosed in the Cited Invention used also have fluorescent properties, there is no ground to approve that the film-like shaped nanocomposite materials disclosed therein are luminous, namely, the films are the "sheet-like illuminants". Therefore, it cannot be said that, even when nanocomposite materials and films are produced without having the objective of obtaining luminous sheets, silicon particles, whose handling is sensitive and the fluorescent properties are easily lost, maintain the stability without being subject to any adverse influence concerning the fluorescent properties.

(2) However, as described above, since the Cited Invention is aiming at improving the stability (luminous stability) of quantum dots, and the problem was solved by surface



improvements of quantum dots (refer to 2(1) described above), as long as the surface modified layers are sound, even after being molded into sheet-like (film-like) shapes, it is reasonable to think that the luminous stability is maintained (Refer to 4(2)A described above. Actually, by referring to the References 1 through 3 presented in 4(2)A described above, even when being molded into sheet-like shapes, the emission characteristics of luminous particles are not lost.). Although the Cited Publication certainly presents various usages utilizing the nonlinearity of index of refraction as application examples to optical devices of quantum dots, as described in 4(2)C, since the Cited Publication focuses attention also on the linear optical properties of quantum dots and mentions about usages in which quantum dots themselves such as optically excited quantum dot lasers emit light, it is not appropriate to think that composite materials formed by dispersing quantum dots in matrix materials and molding into film-like shapes (sheet-like shapes) always lose fluorescent properties, it is rather reasonable to understand that the Cited Invention assumes even application examples using emission stability of quantum dots judging from the problem to be solved.

(3) Therefore, the arguments of the appellant cannot be adopted.

## 6 Summary

As examined above, since it can be said that matters specifying the invention of the Invention related to different features 1, 2 are either what the Cited Invention assumes, or, even if that is not the case, they are somethings that is nothing more than what a person skilled in the art can easily conceive, the Invention is the invention described in the Cited Publication, or is something that could be easily invented by a person skilled in the art on the basis of the Cited Invention.

## No. 5 Conclusion

As explained above, the invention related to Claim 1 of the application is either the invention described in the Cited Publication, or is something that could be easily invented by a person skilled in the art on the basis of the Cited Invention, which corresponds to Article 29(1) of the Patent Act, or is something for which the appellant should not be granted a patent under the provisions of Article 29(2) of the Patent Act.

Therefore, the application shall be refused even without examining the inventions related to the other claims of the application.

Therefore, the appeal decision shall be made as described in the conclusion.

August 17, 2015

Chief administrative judge: KUNISHIMA, Akihiro  
Administrative judge: HIBINO, Takaharu  
Administrative judge: TOYONAGA, Shigehiro