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<td>Inventor(s)</td>
<td>Yam, Vivian Wing-Wah; Au, Vonika Ka-Man; Chan, Mei-Yee; Wong, Keith Man-Chung</td>
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LUMINESCENT GOLD(III) COMPOUNDS FOR ORGANIC LIGHT-EMITTING DEVICES AND THEIR PREPARATION

Inventors: Vivian Wing-Wah YAM, Hong Kong (CN); Vonika Ka-Man AU, Hong Kong (CN); Mei-Yee CHAN, Hong Kong (CN); Keith Man-Chung WONG, Hong Kong (CN)

Correspondence Address:
SALIWANCHIK LLOYD & SALIWANCHIK A PROFESSIONAL ASSOCIATION
PO Box 142950
GAINESVILLE, FL 32614 (US)

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ABSTRACT
A class of luminescent gold(III) compounds containing a tridentate ligand with one strong σ-donating group having the chemical structure depicted by generic formula (I):

\[
\begin{array}{c}
A \\
B \quad N \\
C
\end{array}
\]

wherein:
(a) X is selected from nitrogen;
(b) Y and Z selected from carbon;
(c) A is cyclic structure derivative of pyridine group;
(d) B and C are cyclic structure derivative of phenyl groups;
(e) R₂ is an optionally substituted carbon donor ligand attached to the gold atom;
(f) n is zero, a positive integer or a negative integer.

Related U.S. Application Data
Continuation-in-part of application No. 10/977,200, filed on Oct. 29, 2004, now Pat. No. 7,572,912.
FIG. 5

FIG. 6
ITO/NPB[70]/CBP[5]/6% compound 1 : CBP[30]/BAIq[30]/LiF[0.5]/Al[100]

FIG. 7

Normalized EL Intensity

1.0
0.6
0.2
0.0

350 450 550 650 750

Wavelength (nm)

1% 2% 4% 6%

FIG. 8
FIG. 9
LUMINESCENT GOLD(III) COMPOUNDS FOR ORGANIC LIGHT-EMITTING DEVICES AND THEIR PREPARATION

CROSS-REFERENCE TO RELATED APPLICATION

[0001] This application is a continuation in part of U.S. application Ser. No. 10/977,200, filed Oct. 29, 2004, which is hereby incorporated by reference in its entirety.

FIELD OF THE INVENTION

[0002] The present invention is related to the design and synthesis of novel classes of gold(III) compounds containing a tridentate ligand with one strong d-donating group. Such compounds can be used as light-emitting material in phosphorescence-based organic light-emitting devices (OLEDs).

BACKGROUND

[0003] In recent years, much attention has been drawn towards the research and development of organic light-emitting devices. Such enormous increase in research interest is highly correlated to the potential application of OLEDs in commercial flat panel displays. With the advantages of low cost, light weight, low operating voltage, high brightness, robustness, color tunability, wide viewing angle, ease of fabrication onto flexible substrates as well as low energy consumption, OLEDs are considered as remarkably attractive candidates for flat panel display technologies.

[0004] Typically an OLED contains several layers of semiconductor sandwiched between two electrodes. The cathode is composed of a low work function metal alloy deposited by vacuum evaporation, whereas the anode is a transparent conductor such as indium tin oxide (ITO). Upon the application of a DC voltage, holes injected by the ITO electrode and electrons injected by the metal electrode will recombine to form excitons. Subsequent relaxation of excitons will then result in the generation of electroluminescence (EL).

[0005] In order to achieve higher OLED performance, multiple organic semiconductor layers can be incorporated to further separate the two electrodes. There are two main categories of these semiconductor layers, namely vacuum-deposited small molecules and spin-coated polymeric materials. Both fabrication methods have their respective advantages. Vacuum deposition can allow better control over layer thickness and uniformity, while spin coating offers the ease of use and lower production cost [Burrows, P. E.; Forrest, S. R.; Thompson, M. E. Current Opinion in Solid State and Materials Science, 236 (1997)].

[0006] In spite of the fact that electroluminescence from organic polymers was initially reported in the 1970s [Kaneto, K.; Yoshino, K.; Koo, K.; Imushiki, Y. Jpn. J. Appl. Phys., 1023 (1974)], it was only after the report on yellow-green electroluminescence from poly(p-phenylenevinylene) (PPV) that light-emitting polymers and OLEDs have received much attention [Burroughs, J. H.; Bradley, D. D. C.; Brown, A. R.; Marks, N.; Friend, R. H.; Burn, P. L.; Holmes, A. B. Nature 347, 530 (1990)]. Later on, similar studies were reported by using PPV derivatives as the light-emitting polymers [Brown, D.; Heeger, A. J. Appl. Phys. Lett. 58, 1982 (1991)]. Since then a number of new electroluminescent polymers have been investigated for improved properties.

[0007] Electroluminescence of organic materials was discovered in anthracene crystals immersed in liquid electrolyte in 1965 [Helfruch, W.; Schneider, W. G. Phys. Rev. Lett. 14, 229 (1965)]. Although lower operating voltages could be achieved by using a thin film of anthracene as well as solid electrodes, very low efficiency of such a single-layer device was encountered. High-performance green electroluminescence from an organic small molecule, aluminum tris(quinolate) (Alq3), was first reported in 1987 [Tang, C. W.; VanSlyke, S. A. Appl. Phys. Lett. 51, 913 (1987)]. A double-layer OLED with high efficiency and low operating voltage was described, in which Alq3 was utilized both as emitting layer and electron transporting layer. Subsequent modifications of the device with triple-layer structure gave better performance with higher efficiency.

[0008] For best performance of phosphorescence-based OLEDs, it is desirable for the semiconducting materials to have short lifetimes. One way is to mix singlet and triplet excited states by making use of spin-orbit (L-S) coupling. In the presence of a heavy metal center, the chance of spin-orbit coupling can be greatly enhanced. Hence, the use of heavy metal complexes in OLEDs is preferred over purely organic materials, in which the lowest energy excited state of an organometallic compound is commonly a metal-to-ligand charge transfer (MLCT) triplet state, mixed with the excited singlet state through L-S coupling, so as to obtain higher photoluminescence efficiencies [Baldo, M. A.; Thompson, M. E.; Forrest, S. R. Pure Appl. Chem. 71, 2095 (1999)]. In 1998, Baldo et al. demonstrated a phosphorescence electroluminescence device with high quantum efficiency by using platinum(II) 2,3,7,8,12,13,17,18-octacetyl-21H,23H-porphine (POEP) as a dye. [Baldo, M. A.; O'Brien, D. F.; You, Y.; Shoustikov, A.; Sibley, S.; Thompson, M. E.; Forrest, S. R. Nature 395, 151 (1998)]. A multilayer device in which the emitting layer of Alq3 is doped with POEP showed a strong emission at 650 nm attributed to the triplet excitons of POEP. Cyclometalated iridium(III) is known to show phosphorescence and is another class of materials used for high efficiency OLEDs. Baldo et al. reported the use of fac-tri(2-phenylpyridine)iridium(III) [Ir(ppy)3] as phosphorescence emitting material which was doped in 4,4′-N,N′-diarylazobiphenyl (CBP) as a host in an OLED to give high quantum efficiency [Baldo, M. A.; Lamansky, S.; Burrows, P. E.; Thompson, M. E.; Forrest, S. R. Appl. Phys. Lett. 75, 4 (1999)]. In addition, fac-tri(phenylpyridine)iridium(III) [Ir(ppy)3] was used as phosphorescence sensitizer for high efficiency fluorescent OLED [Baldo, M. A.; Thompson, M. E.; Forrest, S. R. Nature 403, 750 (2000)]. Using the concept of a phosphorescence emitter with a higher population of excitons, very high efficiency red electroluminescence from [2-methyl-6-[2-(2,3,6,7-tetrahydro-1H-5H-2-benz[H]quinolinizin-9-yl)-ethenyl]-4H-pyran-4-ylidene]propanedinitrile (DCM) was found in a multilayer OLED composed of Ir(ppy)3, and DCM dopant layers. In a sensitization process, energy is transferred from Ir(ppy)3 to DCM to give such high efficiency electroluminescence.

[0009] Apart from the enhancement of the emission efficiency, the ability to bring about a variation in the emission color would be important. Most of the common approaches involve the use of different emission characteristics for color tuning. Examples that employ a single light-emitting material as dopant to generate more than one emission color have been rare. Recent studies have shown that different emission colors from a single emissive dopant could be generated by using phosphorescent material through a change in the direction of the bias or in the dopant concentration. Weller et al. reported the fabrication of a simple OLED consisting of semiconduct-
ing polymer PPV and phosphorescent ruthenium polypyridine dopant [Weller, S.: Krumm, K.; Hofstraan, J. W.; De Cola, D. Nature 421, 54 (2003)]. At forward bias, red emission from the excited state of the phosphorescent ruthenium polypyridine dopant was observed, while the OLED emitted a green emission at reverse bias that in the lowest excited singlet state of PPV was populated. Adamovich et al. reported the use of a series of phosphorescent platinum(II) [2-(4,6-difluorophenyl)pyridinato-N,C] β-diketones as single emissive dopant in OLED [Adamovich, V.; Brooks, J.; Tamayo, A.; Alexander, A. M.; Djurovic, P. R.; D’Andrade, B. W.; Adachi, C.; Forrest, S. R.; Thompson, M. E. New J. Chem. 26, 1171 (2002)]. Both blue emission from monomeric species and orange emission from the aggregates were observed in such OLED and the relative intensity of the orange emission increases as the doping level is increased. As a result, the electroluminescence color can be tuned by changing the dopant concentration with equal intensities of the monomeric and aggregate bands. In both cases, the change of electroluminescence color in OLED can be accomplished upon a variation of the external stimulus or fabrication conditions while keeping the light-emitting material the same.

Even though there has been an increasing interest in electrophosphorescent materials, particularly metal complexes with heavy metal centers, most of the work has been focused on the use of iridium(III), platinum(II) and ruthenium (II), whereas the use of other metal centers have been much less explored. In contrast to the isoelectronic platinum(II) compounds which are known to show rich luminescence properties, very few examples of luminescent gold(III) complexes have been reported, probably due to the presence of low-energy d-d ligand field (LL) states and the electrophilicity of the gold(II) metal center. One way to enhance luminescence of gold(III) complexes is through the introduction of strong σ-donating ligands, which was first demonstrated by Yam et al. in which stable gold(III) aryl compounds were synthesized and found to display interesting photoluminescence properties even at room temperature [Yam, W. W.; Choi, S. K.; Lai, T. F.; Lee, W. K. J. Chem. Soc., Dalton Trans. 1001 (1993)]. Another interesting donor ligand is the alkynyl group. Although the luminescence properties of gold(I) alkynyls have been extensively studied, the chemistry of gold(III) alkynyls has been essentially ignored, except for a brief report on the synthesis of an alkynylgold(III) compound of 6-benzyl-2,2’-bipyridine in the literature [Cinellu, M. A.; Minghetti, G.; Pinu, M. V.; Stoccoiro, S.; Zucchi, A.; Manussero, M. J. Chem. Soc. Dalton Trans 2823 (1999)], but their luminescence behaviour has remained totally unexplored. Yam et al. later synthesized a series of bis-cyclometalated alkynylgold(III) compounds using various strong σ-donating alkynyl ligands, and all these compounds were found to exhibit rich luminescence behaviors at both room and low temperatures in various media [Yam, W. W.; Wong, K. M.-C.; Hung, L.-L.; Zhu, N. Angew. Chem. Int. Ed. 44, 3107 (2005); Wong, K. M.-C.; Hung, L.-L.; Lam, W. H.; Zhu, N.; Yam, W. W.-J. Am. Chem. Soc. 129, 4350 (2007); Wong, K. M.-C.; Zhu, X.; Hung, L.-L.; Zhu, N.; Yam, W. W.-W.; Kwok, H. S. Chem. Commun. 2906 (2005)].

Brief Summary

The present invention is directed to novel luminescent cyclometalated gold(III) compounds, their preparation, and OLEDs containing them. Described below is a report of the design and synthesis of various novel gold(III) compounds.

The objective of the present invention is to provide a novel class of cyclometalated gold(III) compounds for applications in organic light-emitting devices. The invented gold(III) compound is a coordination compound that contains a cyclometalated ligand and at least one strong σ-donating group, both coordinated to a gold(III) metal center. Likewise, any σ-donating group can be employed as the ancillary ligand, for instance, alkynyl groups.

The compounds have the chemical structure shown in generic formula (I),

![Diagram](image)

wherein:

(a) X is selected from nitrogen;

(b) Y and Z selected from carbon;

(c) A is cyclic structure derivative of pyridine group;

(d) B and C are cyclic structure derivative of phenyl groups;

(e) R is an optionally substituted carbon donor ligand attached to the gold atom;

(n is zero, a positive integer or a negative integer.

It is expected that the luminescent gold(III) compounds of the present invention will show strong photoluminescence via triplet excited state upon photo-excitation, or electroluminescence via triplet excitation upon applying a DC voltage. Such luminescence properties are observed in compounds that have been synthesized. Preferred compounds of the invention are thermally stable and volatile enough to be able to form a thin layer by sublimation or vacuum deposition. Alternatively, the compounds can be doped into a host matrix for thin film deposition by spin-coating or inkjet printing or other known fabrication methods.

The present invention is also directed to the use of luminescent compounds of general formula (I) as phosphorescent emitters or dopants for the fabrication of OLEDs to generate electroluminescence. Some examples of this class of compounds are illustrated in the present invention for the fabrication of OLEDs with improved current and power efficiencies.

In an OLED according to the present invention, the luminescent gold(III) compound is included in a light-emitting layer. The typical structure of an OLED using luminescent compounds of the present invention as a light-emitting layer is in the order shown in FIG. 1: cathode/electron transporting layer/luminescent gold(III) compound as a light-emitting layer/carrier refinement layer/hole transporting layer/anode.

Brief Description of the Drawings

FIG. 1 is a schematic diagram of the basic structure of an organic EL device.
FIG. 2 shows the UV-vis absorption spectrum of compound 2 in dichloromethane at 298 K. FIG. 3 shows the UV-vis absorption spectra of compounds 3 and 5 in dichloromethane at 298 K. FIG. 4 shows the emission spectrum of compound 2 in dichloromethane at 298 K. No instrumental correction was applied for the emission wavelength. FIG. 5 shows the emission spectra of 5 in degassed dichloromethane at 298 K. No instrumental correction was applied for the emission wavelength. FIG. 6 shows the external quantum efficiency of device with the structure of ITO/NPB (70 nm)/6% compound 1:CBP (30 nm)/BAq (30 nm)/LiF (0.8 nm)/Al (80 nm). FIG. 7 shows the external quantum efficiency of device with the structure of ITO/NPB (70 nm)/CBP (5 nm)/6% compound 1:CBP (30 nm)/BAq (30 nm)/LiF (0.8 nm)/Al (80 nm), in which the CBP layer is used as the carrier confinement layer. FIG. 8 shows the electroluminescence spectra of device of Example 3. FIG. 9 shows a) current efficiency and b) power efficiency of devices of Example 3.

DETAILED DESCRIPTION

The present invention is directed to the synthesis and luminescence studies of a class of luminescent gold(II) compounds with one tridentate ligand and one strong σ-donating group; and the use of such compounds as light-emitting material in OLEDs to provide electroluminescence with high efficiency and brightness. The compounds have the chemical structure shown in generic formula (I),

wherein:
(a) X is selected from nitrogen;
(b) Y and Z selected from carbon;
(c) A is cyclic structure derivative of pyridine group;
(d) B and C are cyclic structure derivative of phenyl groups;
(e) R, is an optionally substituted carbon donor ligand attached to the gold atom;
(f) (I) is the charge on the compound and is zero for a neutral compound or can be a positive integer or a negative integer where the compound is the cation or anion, respectively, of a salt. R, is selected from, but is not limited to, alkylalkyl, substituted alkylalkyl, aryalkyl, substituted aryalkyl, heteroaryalkyl and substituted heteroaryalkyl.

Ring A is pyridine or pyridyl derivative, but is not limited to, with one or more alkyl, alkenyl, alkynyl, alkylaryl, cycloalkyl, OR, NR, SR, C(O)R, C(O)OR, C(O)NRR, CN, CF, NO, SO, SO, hal, aryl, substituted aryl, heteroaryl, substituted heteroaryl or heterocyclic group. Rings B and C are benzene or phenyl derivatives, but are not limited to, with one or more alkyl, alkenyl, alkynyl, alkylaryl, cycloalkyl, OR, NR, SR, C(O)R, C(O)OR, C(O)NRR, CN, CF, NO, SO, SO, hal, aryl, substituted aryl, heteroaryl, substituted heteroaryl or heterocyclic group.

In the present disclosure the following terms are used.

The term “optional” or “optionally” means that the subsequently described event or circumstance may or may not occur, and that the description includes instances where said event or circumstance occurs and instances in which it does not. For example, “optionally substituted alkyl” includes “alkyl” and “substituted alkyl,” as defined below.

The term “halo” or “halogen” as used herein includes fluorine, chlorine, bromine and iodine. The term “alkyl” as used herein includes straight and branched chain alkyl groups, as well as cycloalkyl group with cyclic structure of alkyl groups. Preferred alkyl groups are those containing from one to eighteen carbon atoms and includes methyl, ethyl, propyl, isopropyl, butyl, isobutyl, tert-butyl, and the like. In addition, the alkyl group may be optionally substituted with one or more substituents selected from OR, NR, SR, C(O)R, C(O)OR, C(O)NRR, CN, CF, NO, SO, SO, hal, and cyclic-amino.

The term “alkenyl” as used herein includes both straight and branched chain alkene radicals. Preferred alkenyl groups are those containing two to eighteen carbon atoms. In addition, the alkenyl group may be optionally substituted with one or more substituents selected from OR, NR, SR, C(O)R, C(O)OR, C(O)NRR, CN, CF, NO, SO, SO, hal, and cyclic-amino.

The term “aryalkyl” as used herein includes an alkyl group which has an aromatic group as a substituent. In addition, the aryalkyl group may be optionally substituted with one or more substituents selected from OR, NR, SR, C(O)R, C(O)OR, C(O)NRR, CN, CF, NO, SO, SO, hal, and cyclic-amino.

The term “aryalkyl” as used herein includes an alkyl group which has an aromatic group as a substituent. In addition, the aryalkyl group may be optionally substituted with one or more substituents selected from OR, NR, SR, C(O)R, C(O)OR, C(O)NRR, CN, CF, NO, SO, SO, hal, and cyclic-amino.

Preferred alkyl groups are C, through C alkyls. Similarly C through C alkox and aryl groups are preferred. C through C heteroaryl, alkylamino, arylamino, alkylsulfido, arylsulfido, alkylphosphino or arylphosphino groups are preferable.

Aryl alone or in combination includes carbocyclic aromatic systems. The systems may contain one, two or three rings wherein each ring may be attached together in a pendant manner or may be fused. Preferably the rings are 5- or 6-membered rings. Aryl groups include, but are not exclusive to, unsubstituted or substituted derivatives of benzene, naphthylene, pyrene, anthracene, pentaacene, benzo[a]pyrene, chrysene, coronene, coronulene, naphthalene, phenanthrene, triphenylenylene, ovalene, benzo[ghi]perylene, anthanthrene, pentaene, picene, dibenz[a,j]pyrene, benzo[3,4; 9,10]pyrene, benzo[3,4]pyrene, dibenzo[3,4,8,9]pyrene,
dibenz(3,4:6,7)pyrene, dibenzo[1,2,3,4]pyrene, and naphtho[2,3,4]pyrene; wherein the positions of attachment and substitution can be at any carbon of the group that does not sterically inhibit formation of compound of formula (I), as can be appreciated by one skilled in the art. Preferred substituents include but are not limited to alkyl, alkoxy, aryl.

Heteroaryl alone or in combination includes heterocyclic aromatic systems. The systems may contain one, two or three rings wherein each ring may be attached together in a pendant manner or may be fused. Preferably the rings are 5- or 6-membered rings. Heterocyclic and heterocycle refer to a 3 to 7-membered ring containing at least one heteroatom. Heteroaryl groups include, but are not exclusive to, unsubstituted or substituted derivatives of pyridine, thiophene, furan, pyrazole, imidazole, oxazole, isoxazole, thiazole, isothiazole, pyrole, pyrazine, pyridazine, pyrimidine, benzimidazole, benzofuran, benzothiazole, indole, triazole, tetrazole, pyran, thiopyran, oxadiazole, triazine, tetrazine, carbazole, dibenzo[b]thiophene, dibenzofuran, isodindole, quinoline, isoquinoline, chromene, isochromene and non-aromatic rings including but not limited to pipergasoline, pyrrole, and pyridine; wherein the position of attachment or substitution can be at any carbon of the group that does not sterically inhibit formation of compound of formula (I), as can be appreciated by one skilled in the art. Preferred substituents include but are not limited to alkyl, alkoxy, aryl.

Heteroatom refers to S, O, N, P.

The cyclic structure derivative of pyridine group A is attached at the 2 and 6 position of the pyridine ring to B and C. The group A can be substituted in the 3, 4 or 5 positions. Appropriate substituents at the 3 and 4 are those of appropriate size to allow simultaneous bonding of rings A, B and C, B and C are cyclic structure derived from phenyl groups and can be substituted in any manner that permits simultaneous Au—C and Au—N bonds between the Au atom and A, B, C and R, as compound (I). B and C can be polyaromatics, such as naphthalene units, for example. The rings A and B can be combined as a pyridine and a phenyl derived portion of a polyaromatic heterocyclic structure. For example A and B can be provided from, for example, 7,8-benzquinodimine or other polyaromatic heterocyclic where appropriate chemical bonding to the Au atom permits formation of the gold(III) compound (I). In like manner all three rings, A, B and C can be provided from, for example, dibenzoc[2,1,3]oxadizine or other polyaromatic heterocyclic where appropriate chemical bonding to the Au atom permits formation of the gold(III) compound.
instance, a series of gold(III) aryl complexes was found to show interesting luminescence properties even at room temperature and were stable upon light irradiation [Yam et al., J. Chem. Soc. Dalton Trans. 100 (1993)]. Later on, a class of luminescence bisacylometalated alkynylgold(III) complexes have been reported, in which strong σ-donating alkynyl groups have been incorporated into the gold(III) metal center to improve the emission properties. These complexes were found to be emissive at both room and low temperatures in various media [Yam et al. Angew. Chem. Int. Ed. 44, 3107 (2005); Wong et al. J. Am. Chem. Soc. 129, 4350 (2007)]. The use of such alkynylgold(III) compounds as electrophosphorescent emitters or dopants of organic light-emitting diodes (OLEDs) has recently been communicated in the literature [Wong et al. Chem. Commun. 2906 (2005)].

[0055] In addition to those gold(III) complex with tetrade- nate cyclometalated ligands reported in US Pat. No. 2006/ 0091378 A1, the present invention will also employ various new gold(III) compounds as the light-emitting material for OLEDs. Furthermore, the design and composition of the organic electro-luminescent device has been modified in the present invention, resulting in much improved OLED performance in terms of external quantum efficiencies.

[0056] The luminescent gold(III) compounds of the present invention can be formed into thin films by vacuum deposition, spin-coating, inkjet printing or other known fabrication methods. Different multilayer OLEDs have been fabricated using the compounds of the present invention as light-emitting material or as dopant in the emitting layer. In general, the OLEDs consist of an anode and a cathode, between which are the hole transporting layer, light-emitting layer, and electron transporting or injection layer. The present invention makes use of an additional carrier confinement layer to improve the performance of the devices.

[0057] Referring to FIG. 1, an organic EL device has, in order, a substrate, a hole-injecting anode, a hole transporting layer, a carrier confinement layer, a light-emitting layer, an electron transporting layer, and an electron-injecting cathode.

[0058] The substrate is electrically insulated and can be either optically transparent (e.g., glass or plastic foil) or opaque (e.g., semiconducting materials or ceramics). For viewing the EL emission through the substrate, or through both sides of the device, a glass substrate or a plastic foil is used. For applications where the EL emission is viewed through the top electrode, opaque semiconductor and ceramic wafers can be used.

[0059] The hole-injecting anode injects holes into the organic EL layer when this anode is positively biased. The anode is composed of a conductive and optionally transmissive layer. If the viewing of the EL emission through the substrate is desirable, the hole-injecting anode should be transparent. For applications where the EL emission is viewed through the top electrode, the transmissive characteristic of the anode is immaterial, and therefore any appropriate materials such as metals or metal complexes having a work function of greater than 4.3 eV can be used. Possible metals include gold, indium, molybdenum, palladium, and platinum. If the anode should be transmissive, suitable materials can be selected from the group of metal oxides, including indium-tin oxide, aluminum- or indium-doped zinc oxide, tin oxide, magnesium-indium oxide, nickel-tungsten oxide, and cadmium-tin oxide. The desired metals and metal oxides can be deposited by evaporation, sputtering, laser ablation, and chemical vapor deposition.

[0060] The hole-transporting layer contains at least one hole-transporting aromatic tertiary amine. Suitable materials for use in the hole-transporting layer include 4,4′-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB), 4,4′-bis[N-(3-methylphenyl)-N-phenylamino]biphenyl (TDP), 4,4′,4″-tris[(3-methylphenyl)phenylamino]triphenylamine (MDTDA), and di-4-(N,N-dimethylamino)phenylcyclohexane (TAPC). The carrier confinement layer is used to prevent the migration of triplet excitons out of the light-emitting layer. The carrier confinement layer should have the following features: i) the triplet energy must be sufficiently high to suppress the Dexter energy transfer from the phosphorescent triplet state of dopant material to a lower-lying, nonradiative triplet state of the hole-transporting material; ii) the hole-transporting properties and the highest occupied molecular orbital (HOMO) energy level should facilitate hole injection from the hole-transporting layer to the light-emitting layer; iii) the lowest unoccupied molecular orbital (LUMO) energy level should be above that of the host material in the light-emitting layer. Preferred materials for use in forming the carrier confinement layer are 4,4′-bis(carbazol-9-yl)biphenyl (CBP), m-(N,N-di-carbazole)benzene (mCz) and 4,4′,4″-tris(carbazol-9-yl)triphenylamine (TCTA). The light-emitting layer is formed by doping the phosphorescent Au(III) metal complex as a dopant into a host compound. Suitable host materials should be selected so that the triplet exciton can be transferred efficiently from the host material to the phosphorescent dopant material. Suitable host materials include certain aryl amines, triazoles and carbazole compounds. Examples of desirable hosts are CBP, mCz, TCTA, 3-(4-hydroxyphenyl)-4-phenyl-5-tetralonylephyl-1,2,4-triazole (TAZ), p-bis(triphenylisilyl)benzene (UG12), and poly(N-vinylcarbazole) (PVK). The electron-transporting layer consists of materials or mixtures of materials which have a high ionization potential and wide optical band gap. Suitable electron transporting materials include 1,3,5-tris(phenyl-2-benzimidazolyl)-benzene (TPBI), bathocuproine (BCP), batho-phenanthroline (Bphen) and bis(2-methyl-8-quinolinolato)-4-(phenylphenolate)aluminum (BA1q). The organic film can be prepared by thermal evaporation, spin-coating, inkjet printing from a solution, or other known fabrication methods.

[0061] The top-electron-injecting cathode acts as a trans- missive electron injector that injects electrons into the organic EL layer when the cathode is negatively biased. The cathode is formed by a thin fluoride layer (which may be omitted) and a metal or metal alloy, preferably having a work function of less than 4 eV. Suitable materials include Mg:Ag, Li:Ag, Al.

[0062] The present invention will be illustrated more specifically by the following non-limiting examples. It being understood that changes and variations can be made therein without deviating from the scope and the spirit of the invention as hereinafter claimed. It is also understood that various theories as to why the invention works are not intended to be limiting.

[0063] The compounds described above have been represented throughout by their monomeric structure. As is well known to those in the art, the compounds may also be present as dimers, trimers, oligomers, polymers or dendrimers. For example, R1 can be a donor ligand with a structure where two Au—C bonds can be formed between two different C atoms of R1 and two different Au atoms. Alternately substituents can be bridging groups between two different rings A, B, or C.
between one of the rings and $R_i$ where two or more monomeric structures are combined into a dimeric, trimeric oligomeric or polymeric structure.

**Example 1**

**General Synthetic Methodology**

![Chemical Structure](image)

wherein:

- **[0065]** (a) $R_i$ is selected from, but not limited to, alkylalkynyl, substituted alkylalkynyl, arylalkynyl, substituted arylalkynyl, heteroarylalkynyl, substituted heteroarylalkynyl and $(C=)$, where $(C=)$ represents a carbon-carbon triple bond, $n=1-8$, and $R_{13}$ is selected from alkyl, aryl, substituted aryl, heteroaryl, substituted heteroaryl and tri (alkyl)dialkyl.
- **[0066]** (b) $R_2-R_{12}$ groups are each independently selected from, but is not limited to, hydrogen, alkyl, alkenyl, alkynyl, alkylalkynyl, arylalkynyl, substituted arylalkynyl, alkenyl, alkynyl, alkylalkynyl, alkylalkynyl, alkylalkynyl, OR, NR, SR, CO, COOR, CN, CF, NO, SO, SOR, SO$_2$R, halo, aryl, substituted aryl, heteroaryl, substituted heteroaryl or a heterocyclic group.

As can be appreciated by those skilled in the art, the position, structure and size of the substituents $R_2$ through $R_{12}$ and any substituents included on $R_i$ are chosen to avoid steric inhibition of Au—C bond formation. In some embodiments of the invention $R_2$ and $R_3$ and/or $R_5$ and $R_6$ can be combined into a bridging group of two or more atoms that permit the planes of the pyridyl and phenyl rings to achieve the proper orientation for formation of the Au—C and Au—N bonds of compound (II).

**Example 2**

**Synthesis and Characterization**

**[0067]** Compounds 1, 2, 7 and 11 have been reported in the literature [Wong et al. *Chem. Commun.* 2906-2908 (2005); Wong et al., *J. Am. Chem. Soc.* 129, 4350 (2007)]. Compounds 3-6 and 8-10 were synthesized by a similar methodology, in which the tridentate ligands, 2,5-F$_2$Ph-C’N’C and FC’N’CF, and the precursor compounds, [Au(2,5-F$_2$Ph-C’N’C)] and [Au(FC’N’CF)], were respectively prepared according to modified literature procedures [Kuhnt, *F. Synthesis* 1 (1976); Wong, K. H.; Cheung, K. K.; Chan, M. C. W.; Che, C. M. *Organometalics* 17, 5505 (1998)].
The characteristic spectroscopic properties of compounds 1-11 are as follows:

[20068] 80% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 7.01-7.07 (m, 4H, C-C₂H₅ and NPh₂), 7.10-7.13 (m, 4H, NPh₂), 7.26-7.30 (m, 6H, phenyl of C'N'C and NPh₂), 7.40 (dt, J=7.3 and 1.2 Hz, 2H, phenyl of C'N'C), 7.46 (d, J=8.7 Hz, 2H, C=C₂H₅), 7.54 (d, J=8.0 Hz, 2H, pyridyl of C'N'C), 7.62 (dd, J=7.3 and 1.2 Hz, 2H, phenyl of C'N'C), 7.91 (t, J=8.0 Hz, 1H, pyridyl of C'N'C), 8.05 (dd, J=7.3 and 1.2 Hz, 2H, phenyl of C'N'C) Positive FAB-MS: m/z 694 [M⁺]. IR (KBr disc): 2149 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 62.34; H, 3.55; N, 3.86. Caled for C₃₀H₁₇AuN₂O₂: C, 62.36; H, 3.32; N, 3.93.

[20069] 80% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 7.01-7.07 (m, 4H, C=C₂H₅ and NPh₂), 7.10-7.13 (m, 4H, NPh₂), 7.26-7.30 (m, 6H, phenyl of C'N'C and NPh₂), 7.40 (dt, J=7.3 and 1.2 Hz, 2H, phenyl of C'N'C), 7.46 (d, J=8.7 Hz, 2H, C=C₂H₅), 7.54 (d, J=8.0 Hz, 2H, pyridyl of C'N'C), 7.62 (dd, J=7.3 and 1.2 Hz, 2H, phenyl of C'N'C), 7.91 (t, J=8.0 Hz, 1H, pyridyl of C'N'C), 8.05 (dd, J=7.3 and 1.2 Hz, 2H, phenyl of C'N'C). Positive FAB-MS: m/z 694 [M⁺]. IR (KBr disc): 2149 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 62.34; H, 3.55; N, 3.86. Caled for C₃₀H₁₇AuN₂O₂: C, 62.36; H, 3.32; N, 3.93.

[20070] 28% of 1 H NMR (300 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 7.06 (m, 4H, C=C₂H₅ and NPh₂), 7.10 (d, J=8.7 Hz, 2H, NPh₂), 7.24-7.30 (m, 8H, phenyl of C'N'C and NPh₂), 7.36-7.43 (m, 5H, C=C₂H₅ and phenyl of C'N'C), 7.63 (m, 4H, pyridyl and phenyl of C'N'C), 8.01 (d, J=6.1 Hz, 2H, phenyl of C'N'C). Positive FAB-MS: m/z 806 [M⁺]. IR (KBr disc): 2153 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 62.64; H, 3.33; N, 3.19. Caled for C₄₃H₂₈N₄F₂AuH₂O₂C: C, 62.63; H, 3.54; N, 3.40.

Example 3

[20071] 82% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 7.29 (dt, J=7.4 and 1.3 Hz, 2H, phenyl of C'N'C), 7.35-7.37 (m, 3H, C=C₂H₅ and phenyl of C'N'C), 7.41 (dt, J=7.4 and 1.3 Hz, 2H, phenyl of C'N'C), 7.52 (d, J=8.0 Hz, 2H, pyridyl of C'N'C), 7.54-7.57 (m, 2H, C=C₂H₅), 7.61 (dd, J=7.4 and 1.3 Hz, 2H, phenyl of C'N'C), 7.90-7.96 (m, 3H, phenyl and pyridyl of C'N'C). Positive FAB-MS: 71/2.552 [M⁺]. IR (KBr disc): 2095 and 2198 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 58.67; H, 3.01; N, 2.58. Caled for C₃₄H₂₉N₅Au: C, 58.76; H, 2.90; N, 2.54.

[20072] 88% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 6.87 (s, 3H, CH₃), 7.17 (d, J=8.0 Hz, 2H, C=C₂H₅), 7.21 (dt, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.33 (dt, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.44 (d, J=8.0 Hz, 2H, pyridyl of C'N'C), 7.45 (d, J=8.0 Hz, 2H, C=C₂H₅), 7.51 (dd, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.83-7.88 (m, 3H, phenyl and pyridyl of C'N'C). Positive FAB-MS: m/z 566 [M⁺]. IR (KBr disc): 2080 and 2195 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 59.78; H, 3.24; N, 2.69. Caled for C₃₂H₂ₐN₅Au: C, 59.48; H, 3.21; N, 2.48.

[20073] 32% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 8.29 (dt, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.33-7.39 (m, 3H, C=C₂H₅), 7.41 (dt, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.52 (d, J=8.0 Hz, 2H, pyridyl of C'N'C), 7.54-7.57 (m, 2H, C=C₂H₅), 7.61 (dd, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.91 (dd, J=7.6 and 1.3 Hz, 2H, phenyl of C'N'C), 7.92 (t, J=8.0 Hz, 1H, pyridyl of C'N'C). Positive FAB-MS: m/z 576 [M⁺]. IR (KBr disc): 2066 and 2155 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 60.73; H, 2.99; N, 2.50. Caled for C₃₃H₂₉N₅Au: C, 60.53; H, 2.80; N, 2.43.

[20074] 78% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 6.89 (dt, J=8.7 and 2.7 Hz, 2H, phenyl of C'N'C), 7.32-7.41 (m, 5H, phenyl of C'N'C), 7.56-7.61 (m, 4H, phenyl of C'N'C and C=C₂H₅), 7.65 (dd, J=8.7 and 2.7 Hz, 2H, phenyl of C'N'C), 7.85 (t, J=8.0 Hz, 1H, pyridyl of C'N'C). Positive FAB-MS: 71/2.564 [M⁺]. IR (KBr disc): 2151 cm⁻¹ ν(C≡C). Elemental analyses: Found (%) C, 53.02; H, 2.58; N, 2.65. Caled for C₃₃H₂₉N₅Au: C, 53.30; H, 2.50; N, 2.49.

[20075] 85% of 1 H NMR (400 MHz, CDCl₃, 298 K, relative to Me₄Si): δ 1.39 (s, 18H, Bu₃), 7.35 (m, 5H, C=C₂H₅), 7.46 (d, J=8.0 Hz, 2H, pyridyl of Bu₃C⁷B₇), 7.56 (d, J=8.2 Hz, 2H, phenyl of Bu₃C⁷B₇), 7.60 (dd, J=8.2 and 2.0 Hz, 2H, phenyl of Bu₃C⁷B₇), 7.86 (t, J=8.0 Hz, 1H, pyridyl of Bu₃C⁷B₇), 8.18 (d, J=2.0 Hz, 2H, phenyl of Bu₃C⁷B₇). Positive EIMS: m/z 640 [M⁺]. IR
Elemental analyses: Found (%) C, 61.02; H, 5.08; N, 2.17. Caled for C₃₃H₃₂N₃Au: C, 61.11; H, 5.09; N, 2.16.

[Chem. Soc. 129, 4350-4365 (2007)], whereas the photophysical data of selected compounds are summarized in Table 1.

[0081] As shown in FIG. 2, compound 2 exhibits an intense absorption band at ca. 316 nm and a moderately intense vibronic-structured absorption band at 590-410 nm in dichloromethane at 298 K. The low-energy vibronic-structured absorption band shows vibrational progression splittings of ca. 1250 cm⁻¹, corresponding to the skeletal vibrational frequency of the C=N bond. The low-energy absorptions are assigned as intraligand (IL) π-π* transition of the C=N ligand. 

[0082] As shown in FIG. 3, compounds 3 and 5 show low-energy vibronic-structured absorption bands at ca. 362-412 nm in their electronic absorption spectra in dichloromethane at room temperature. Similar to other mononyl analogues, such low-energy absorption band was tentatively assigned as the metal-perturbed IL [π-π*(RC=N)CR'] transition, involving some charge transfer from the phenyl moiety to the pyridyl unit. It is noteworthy that the electronic absorption spectrum of 5 in dichloromethane featured additional highly vibronic-structured absorption bands at 298-362 nm with vibrational progression splittings of ca. 2020 cm⁻¹, which were typical of the v(C=C) stretching frequency. By comparing the absorption spectrum with that of the free ligand 1-(hexa-1,3,5-triynyl)benzene, the absorptions in 5 were assigned as an admixture of the metal-perturbed IL [π-π*(C=N)] and IL [π-π*(C=C)] transitions.

Example 5

Photoluminescence Properties

[0083] In contrast to most other Au(III) compounds which are non- emissive or only show luminescence at low temperature, the compounds display intense luminescence at 450-665 nm in the solution state at room temperature (Table 1).

[0084] FIG. 4 shows the emission spectrum of compound 2 in dichloromethane at room temperature. In addition to an intense emission band at ca. 450 nm, a broad structureless emission band is observed at ca. 665 nm. With reference to the spectroscopic studies on the emission properties of related compounds, this low-energy structureless band is assigned as derived from an excited state of 

\[ \text{ILCT} \ [\pi-\pi*(C=N)CR'] \text{origin due to the presence of energetically higher-lying } \pi-\pi* \text{bands.} \]

[0085] FIG. 5 shows the emission spectrum of mononuclear gold(III) triynyl complex 5 in dichloromethane at room temperature, in which a highly structured and narrow-bandwidth emission band at ca. 518 nm is observed. The vibrational progression spacings of ca. 2100 cm⁻¹ were characteristic of the v(C=C) stretching states in the ground state, indicating the involvement of the hexatriynyl moiety in the excited state. It was likely that upon increasing the π conjugation of the alkynyl unit in the emissive state, leading to an emission that was predominantly originating from a [II] π-π*(C=C₃) excited state of the hexatriynyl moiety.
TABLE 1

<table>
<thead>
<tr>
<th>Compound</th>
<th>Medium (T/K)</th>
<th>Absorption λ_{max}/nm (ε_{max}/dm^3 mol^-1 cm^-1)</th>
<th>Emission λ_{max}/nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CHCl_3 (298)</td>
<td>320 (38270), 384 (9560), 400 (5565), 416 (5590)</td>
<td>620</td>
</tr>
<tr>
<td>2</td>
<td>CHCl_3 (298)</td>
<td>36 (45915), 390 (7810), 408 (8240)</td>
<td>450, 665</td>
</tr>
<tr>
<td>3</td>
<td>CHCl_3 (298)</td>
<td>312 (24350), 326 (22650), 366 (47190), 384 (51090), 404 (4130)</td>
<td>478, 510, 556, 591</td>
</tr>
<tr>
<td>4</td>
<td>CHCl_3 (298)</td>
<td>310 (21050), 324 (18360), 365 (50250), 383 (4875), 404 (3610)</td>
<td>476, 508, 553, 595</td>
</tr>
<tr>
<td>5</td>
<td>CHCl_3 (298)</td>
<td>298 (12005), 316 (38840), 335 (28500), 326 (15590), 386 (54600), 406 (4520)</td>
<td>518, 584, 667</td>
</tr>
<tr>
<td>6</td>
<td>CHCl_3 (298)</td>
<td>306 (13350), 316 (13068), 366 (33300), 384 (4190), 404 (3876)</td>
<td>475, 505, 545, 585</td>
</tr>
<tr>
<td>7</td>
<td>CHCl_3 (298)</td>
<td>313 (26150), 323 (24460), 372 (6130), 392 (8035), 412 (2465)</td>
<td>484, 514, 548, 593</td>
</tr>
<tr>
<td>8</td>
<td>CHCl_3 (298)</td>
<td>314 (13620), 322 (12435), 376 (4016), 396 (5719), 416 (5470)</td>
<td>486, 516, 556, 595</td>
</tr>
<tr>
<td>9</td>
<td>CHCl_3 (298)</td>
<td>313 (14880), 322 (12810), 374 (3808), 384 (5775), 414 (5675)</td>
<td>487, 520, 564, 636</td>
</tr>
<tr>
<td>10</td>
<td>CHCl_3 (298)</td>
<td>290 (4260), 304 (40780), 330 (25220), 364 (6100), 386 (5120), 406 (3540)</td>
<td>475, 505, 547, 592</td>
</tr>
</tbody>
</table>

Example 6

Construction of an Organic EL Device

[0086] An organic EL device according to an embodiment of the invention was constructed in the following manner:

[0087] a) a transparent anode ITO-coated glass substrate was ultrasonicated in a commercial detergent, rinsed in deionized water, and then dried in an oven. The substrate was further subjected to an UV-ozone treatment for 15 minutes;

[0088] b) the substrate was put into a vacuum chamber, and the chamber was pumped down to 5x10^-6 mbar;

[0089] c) a 70 nm thick NPB hole-transporting layer was deposited on ITO-coated glass substrate;

[0090] d) a 30 nm thick doped CBP light-emitting layer was deposited on CBP layer, in which 6% v/v compound 1 was doped into light-emitting CBP layer;

[0091] e) a 30 nm thick BAQ electron-transporting layer was deposited on doped CBP light-emitting layer;

[0092] f) a 0.8 nm thick LiF and a 80 nm thick Al were deposited on the BAQ layer as electron-injecting cathode.

[0093] All materials were prepared by thermal evaporation from tantalum boats. Deposition rates were monitored with a quartz oscillation crystal and controlled at 0.1-0.2 nm/s for both organic and metal layers. Current density-voltage-luminance characteristics of organic EL devices were measured with a programmable Keithley model 237 power source and a SpectraScan PR 650 photometer under ambient air conditions.

Example 7

Construction of Another Organic EL Device

[0094] The same materials and processing procedures were employed as described in Example 6, except that a 5 nm thick carrier confinement CBP layer was inserted in between the hole transporting layer and the light-emitting layer.

[0095] FIG. 6 and FIG. 7 depict the plot of external quantum efficiencies of devices of Example 6 and 7, respectively. Without the carrier confinement layer, the device shows a maximum external quantum efficiency of 7.4%. On the other hand, a remarkable increase in device performance can be achieved by employing a carrier confinement layer, in which such device shows a maximum external quantum efficiency of 12.8%. This confirms that the carrier confinement layer between the hole transporting layer and the light-emitting layer can effectively suppress the Dexter energy transfer from the phosphorescent triplet state of dopant material to the triplet state of hole-transporting material.

Example 8

Construction of Yet Another Organic EL Device

[0096] An organic EL device according to an embodiment of the invention was constructed in the following manner:

[0097] a) a transparent anode ITO-coated glass substrate was ultrasonicated in a commercial detergent, rinsed in deionized water, and then dried in an oven. The substrate was further subjected to an UV-ozone treatment for 15 minutes;

[0098] b) the substrate was put into a vacuum chamber, and the chamber was pumped down to 5x10^-7 mbar;

[0099] c) a 70 nm thick NPB hole-transporting layer was deposited on ITO-coated glass substrate;

[0100] d) a 5 nm thick CBP carrier confinement layer was deposited on NPB layer;
4. The compound of claim 1, wherein rings B and C are benzene or phenyl derivatives with one or more alkyl, alkenyl, alkynyl, alkylaryl, cycloalkyl, OR, NR₂, SR, C(O)R, C(O)OR, C(O)NR₂, CN, CF₃, NO₂, SO₂, OR, SO₂R, halo, aryl, substituted aryl, heteroary, substituted heteroary or heterocyclic group.

5. The compound of claim 1, wherein one or both of the rings B and C are combined with the ring A as a polycyclic aromatic group.

6. The compound of claim 1, wherein two, three or more of the compounds are connected through a bridging substituent as a dimer, trimer, oligomer or polymer.

7. A compound having the chemical structure represented by the following general formula (II),

![Chemical Structure Image]

wherein:
(a) R₁ is selected from alkylalkynyl, substituted alkylalkynyl, aryalkynyl, substituted aryalkynyl, heteroaryalkynyl, substituted heteroaryalkynyl and (C=C)ₙR₁₅, where (C=C) represents a carbon-carbon triple bond, n=1-8, and R₁ is selected from alkyl, aryl, substituted aryl, heteroary, substituted heteroary and tri(alkyl)silyl;
(b) R₂-R₁₅ groups are each independently selected from hydrogen, alkyl, alkenyl, alkynyl, alkylaryl, aryl and cycloalkyl with one or more alkyl, alkenyl, alkynyl, alkylaryl, cycloalkyl, OR, NR₂, SR, C(O)R, C(O)OR, C(O)NR₂, CN, CF₃, NO₂, SO₂, OR, SO₂R, halo, aryl, substituted aryl, heteroary, substituted heteroary or a heterocyclic group.

8. A compound according to claim 6 wherein the groups R₅ and R₆ and/or R₇ and R₈ are combined into a bridging group of two or more atoms.

9. A compound according to claim 6 wherein the compound can be deposited as a thin layer.

10. A compound according to claim 8 wherein the thin layer can be deposited by sublimation, vacuum deposition, spin-coating, inkjet printing, or other known fabrication techniques.

11. A compound according to claim 6, wherein the compound has photoluminescence properties.

12. A compound according to claim 6, wherein the compound has electroluminescence properties.

13. A compound of claim 6, wherein the compound can be fabricated into a light-emitting device.

14. A compound of claim 6, wherein the compound serves as the light-emitting layer of a light-emitting device.

15. A compound of claim 6, wherein the compound serves as a dopant in the light-emitting layer of a light-emitting device.
16. A compound of claim 15 wherein luminescent energy of the compound varies with the concentration of the dopant.

17. A light-emitting device comprising a layer of the compound of claim 6.

18. A dopant comprising a compound according to claim 6.


20. A light-emitting device of claim 19 wherein a carrier confinement layer that has a triplet energy higher than that of the compound of claim 6 is inserted between the hole-transporting layer and the light-emissive layer.

* * * * *