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European Patent No. 1449238
(Application No. 00932308.0/ Appeal No. T0544/12-3.3.09)
The Trustees of Princeton University and
The University of Southern California ("Proprietors")
Third Party Observations

Dear Sirs:

We lodge herewith a Third Party Observation to this European patent (EP-238). We enclose a copy of each prior art publication relied upon.

On March 21, 2013, the Board of Appeals ("BOA"), in a communication sent pursuant to Article 15(1) of its Rule of Procedures, raised certain issues to be considered at the Oral Proceedings ordered by the BOA on March 18, 2013, to be held on November 21st and 22nd, 2013. The purpose of the BOA's communication is to assist the parties in preparation of the oral proceedings. Based upon our review of this communication, including the BOA's provisional but not binding views expressed Paragraph 5.1 concerning "every organometallic iridium compound" and Paragraph 5.2 concerning "every OLED" and concerning novelty (**54 EPC**) and inventive step (**54 & 56 EPC**) Paragraph 6.1, 6.2, 6.3 and 7, and the record we consider it necessary to make this submission.

For the reasons summarized below, and explained more fully in the attached memorandum, we conclude the following: there existed before the broadest claims of EP-238 were filed papers and patents ("prior art") that destroy the novelty (54 EPC) of or otherwise make non-inventive/obvious (54 & 56 EPC) EP-238's claims, especially in the sense that others could have easily created all of the parts contained in the claims of EP-238 without an invention.

The Proprietors' reply to appeal dated December 18, 2012, seems to contain conspicuous omissions at the heart of this matter. Never addressed are the data in EP-238 itself which show that the quantum efficiency of the referenced organic light emitting devices (OLEDs) varies dramatically with the OLED's architecture, the presence of a barrier layer, and the identity of the host material. Notwithstanding these critical elements, EP-238's broadest claims recite any *heterostructure*, any optional *host material*, and both any *phosphorescent organometallic iridium compound* and any *phosphorescent organometallic osmium compound.* Also never addressed is EP-238's attribution of the critical importance of the barrier layer to the prior art work of O'Brien. (**Reference 28: O'Brien [D19]**).

Proprietors' position in their December 18, 2012, submission seems contrary. On one hand, regarding sufficiency of disclosure (**83 EPC**), Proprietors basically argued that the single example from EP-238 would allow one to make any claimed embodiment without undue burden and *without needing inventive skill.* On the other hand, in rebutting the lack of inventive step,

Proprietors argued that modifying the prior art examples would require *inventive skill* to reach an embodiment of the claimed invention. How can this be? For example, as just noted, O'Brien uses substantially the same basic OLED architecture but incorporates a different dopant, i.e., O'Brien would require a simple substitution to reach EP-238's claimed invention. Prior art shows an OLED using a *phosphorescent organometallic iridium compound* having an oxinoid structure, (**Ref. 14: Prior Iridium OLED Patent [US 4,672,265]**), which the prior art regards as *organometallic*. (**Reference 25: Organometallic Oxinoids Patent [S4]**). Prior art also shows an OLED using a *phosphorescent organometallic osmium compound* and contains instructions to modify its embodiment to make an OLED using a *phosphorescent organometallic iridium compound* that is substantially equivalent to EP-238's exemplified compound. (**Reference 23: Ma [D1]**). Proprietors' claims are not patentable.

EP-238's Claims

EP-238 was granted with 30 claims (**Reference 3a: Original Claims**), which are dividable into two Groups. Group I is directed to an *electroluminescent layer*, and the subject matter of Group I is represented by claim 1:

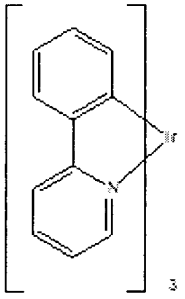
> 1. An *electroluminescent layer* comprising an emissive layer including an emissive molecule that is a phosphorescent organometallic iridium compound or a phosphorescent organometallic osmium compound.

Group II is directed to an *organic light emitting device* including the *electroluminescent layer* from Group I, and the subject matter of Group II is represented by claim 16:

> 16. An *organic light emitting device* comprising a *heterostructure* containing an *emissive layer* that produces *luminescent emission* when a voltage is applied across the heterostructure, wherein the *emissive layer* includes a molecule that is a *phosphorescent organometallic iridium compound* or a *phosphorescent organometallic osmium compound.*

(Reference 1: EP-238, p. 11, l. 48-p. 15, l. 27).

In addition to the representative claims, other claims specify more details about the representative claims and therefore narrow their respective base claim. For example, claim 17 requires, in the OLED, that the emissive layer comprises *a host material and the phosphorescent organometallic compound is present as a guest [i.e., a dopant] in said host material.* Claim 18 requires, in the OLED, that the emissive molecule is *a phosphorescent organometallic iridium compound*, and claim 19 further requires that the *phosphorescent organometallic iridium compound* is $Ir(ppy)_3$, the structure of which is below.



Ir(ppy)$_3$

(**Reference 1: EP-238, p. 13, ll. 41-52**). For reasons that will be apparent, our focus is on the claims from Group II, and the emphasized terms will be elaborated upon with reference to EP-238's specification.

EP-238's Specification

Organic light emitting devices, or "*OLEDs*," as used in the claims and therefore as relevant here, include two or more organic layers between a cathode and an electrode (a *heterostructure*) in which one of the layers contains an organic molecule that can be made to emit light by applying a voltage across the device. (**Reference 1: EP-238, ¶2**). One layer is an electron transport layer (ETL), and the other is a hole transport layer (HTL). Either the ETL or the HTL may emit light and therefore may be the *emissive layer*. Alternatively, another additional layer emits light and therefore is the *emissive layer*. In any case, the *emissive layer* contains a molecule that is either a *phosphorescent organometallic iridium compound* or a *phosphorescent organometallic osmium compound* and optionally *a host material*. Clearly, representative claim 16 is very broad in the sense that the claim language never defines the architecture of the *OLED*, and the claim language tolerates the presence of any *phosphorescent organometallic iridium compound* or any *phosphorescent organometallic osmium compound* and any optional *host material.*

With this extremely broad scope of claim, one would have expected EP-238 to have described a pioneering invention having a lot of examples. Yet upon further examination, and as explained below, others invented the *OLED* architecture, *phosphorescent organometallic iridium compound, phosphorescent organometallic osmium compound*, and *host material*. As explained next, EP-238's broad patent scope is based on testing of a single *phosphorescent organometallic iridium compound*—Ir(ppy)$_3$—in several embodiments of *OLED* architecture and with two different *host materials*.

The first tested *OLED* contained, between the cathode and anode, four separate organic layers. Three of the four layers consisted of an ETL, HTL, and *emissive layer*, i.e., the layer containing Ir(ppy)$_3$ and optionally a *host material* called CBP. The fourth layer seemingly improves, in conjunction with other structure, the efficiency of emitted light and is called a barrier layer (BL).

The first *OLED* was tested to determine the effect of the component parts of the *emissive layer*. In one embodiment (**A**), the *emissive layer* consisted of 100% Ir(ppy)$_3$, and the *OLED* emitted light having a quantum efficiency of about 0.8%. In another embodiment (**B**), the

emissive layer consisted of 94% *host material* (CBP) and 6% of $Ir(ppy)_3$, and the *OLED* emitted light having a quantum efficiency of about 8%. Clearly, the *host material* has a dramatic effect and the mere presence of $Ir(ppy)_3$ in an *emissive layer* is insufficient to make any and all *OLED* architectures have strong emission.

A second test still used the first tested *OLED* and further analyzed the destructive effect of altering the *host material* of embodiment (**B**). While the *host material* of embodiment (**B**) is CBP, the *host material* of embodiment (**C**) is another material called AlQ_3. Having a different *host material*, the second tested *OLED* emitted light having a much lower quantum-efficiency of about 0.2%. Clearly, the identity of the *host material* is essential to the *OLED's* ability to have higher quantum-efficiencies.

A third test determined the destructive effect of removing the barrier layer (BL) from the first tested *OLED* architecture using the *emissive layer* of embodiment (**B**). In other words, the third tested *OLED* has the same architecture as the first tested *OLED*, but the third tested *OLED* lacks the barrier layer (BL). Having no barrier layer (BL), the third tested *OLED* emitted light having a much lower quantum-efficiency of about 0.2%. Clearly, the *OLED* architecture, i.e., the presence of the barrier layer (BL), is essential to the *OLED's* ability to have higher quantum efficiencies. (**Reference 1: EP-238, ¶25**) (admitting that the "barrier layer ... was necessary to ... maintain high efficiencies.").

In short, these data in EP-238 show that the quantum efficiency varies dramatically with the OLED architecture, the presence of a barrier layer (BL), and the identity of the *host material*. Thus, it is surprising that EP-238's broadest claims recite any *heterostructure*, optional *host material*, and *phosphorescent organometallic iridium compound.*

Even more surprising is the fact that EP-238's broadest claims further recite any *phosphorescent organometallic osmium compound.* For support, EP-238 further states its emissive molecules could also include "[o]rganometallic compounds of osmium." (**Reference 1: EP-238, ¶32**). No emissive molecule other than $Ir(ppy)_3$ was tested.

Prior Art

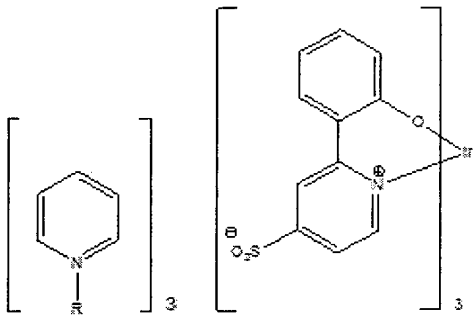
Prior art showing the exemplified OLED architecture of EP-238

EP-238 candidly credits another publication for developing the particular OLED architecture used in its examples. (**Reference 1: EP-238, ¶25**) (crediting D.F. O'Brien, et al., "Improved energy transfer in electrophosphorescent devices," Appl. Phys. Let. 1999, 74, 442-444 (**Reference 28: O'Brien [D19]**)). In particular, O'Brien describes the same OLED heterostructure that was used in Example 2 of EP-238, except for minor variations in the thickness of the individual layers and for the particular emissive molecule. While O'Brien describes using a platinum compound, namely, PtOEP, EP-238 describes using $Ir(ppy)_3$. Clearly, to modify O'Brien's example in a way to reach the embodiment of Example 2 in EP-238 would require a simple substitution of O'Brien's PtOEP with EP-238's $Ir(ppy)_3$.

Prior art showing OLEDs using *phosphorescent organometallic* iridium *compounds*

A Japanese patent, applied for in 1984, titled "Driving Method of EL Element," under Japanese Patent No. (A)H 07-263145 (**Reference 13: JP-145 [S7-S8]**), shows IrQ'_3 on page 7, in

the top-left-most structure in which M=Ir, Q' = pyridium 8-hydroxyquinoline-4-sulfonate. (**Reference 13: JP-145 [S7-S8]**). See to the right.

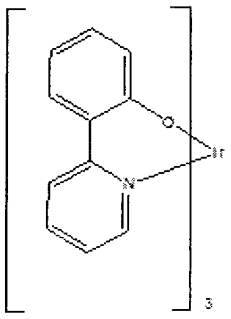


IrQ'_3

JP-145 also shows the use of IrQ'_3 in each luminescent layer of its OLED, e.g., Figure 2. (**Reference 13: JP-145, p. 7; Figs. 2-3 [S7-S8]**).

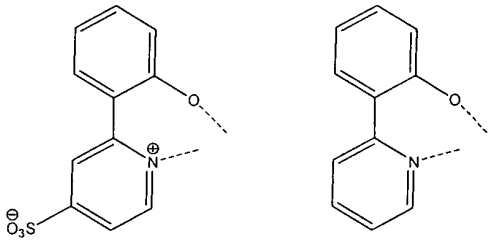
Similarly, IrQ'_3 is also described in U.S. Patent no. 4,672,265 from 1987 (**Reference 14: Prior Iridium OLED Patent [US 4,672,265]**) as an example of an electroluminescent organic compound having high luminescent quantum efficiency. The electroluminescent organic compounds, including IrQ'_3, are inserted into a *heterostructure* having two or more layers (**Reference 14: Prior Iridium OLED Patent [US 4,672,265], col. 10**) between electrodes. (**Reference 14: Prior Iridium OLED Patent [US 4,672,265], col. 15, ll. 24-30; see also Figs. 2-4**).

IrQ'_3 is *a phosphorescent ... iridium compound* as recited in claim 16. IrQ'_3 is closely related in structure to IrQ_3, shown below.



IrQ_3

IrQ_3 and IrQ'_3 are analogs, in which IrQ'_3 has a sulfonate group where IrQ_3 has hydrogen on its ligand.



Q' Q

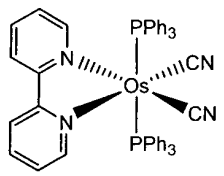
(See the structures above, on the lower ring of each, at 8 o'clock. Notice the $^{-}O_3S$ on Q' v. nothing on Q.). This close structural similarity raises a presumption and expectation that both IrQ'_3 and IrQ_3 possess similar properties. Along these lines, one of the inventors of EP-238 admitted that IrQ_3 is *phosphorescent*. "Organic Light-emitting Devices Based on Phosphorescent Hosts and Dyes" by R.C. Kwong, S. Lamansky, and M. E. Thompson, published in *Advanced Materials* in 2000, refers to the phosphorescence yield of IrQ_3. **(Reference 15: Kwong [S9])**. Since IrQ_3 has a close structural relationship to IrQ'_3, and IrQ_3 is *phosphorescent*, there is no reason to believe that IrQ'_3 is not *phosphorescent*. In any case, IrQ'_3 and IrQ_3 possess similar luminescent properties, which are of course corroborated by both JP-145's and the Prior Iridium OLED Patents' use of IrQ'_3 and Kwong's use of IrQ_3 in OLED.

IrQ'_3 is a member of a class of molecules called oxinoids, which were recognized as a generic class of compounds being highly desirable for OLED and alternatively for use as a dopant or as a host material. U.S. Pat. No. 5,150,006, "Blue Emitting Internal Junction Organic Electroluminescent Device (II)." **(Reference 16: Prior Oxinoid Patent, col. 18, ll. 12-28)**. Thus, there is a good reason to use members of this art-recognized class of emissive molecules that are *organometallic*. U.S. Patent No. 5,486,406, Green-emitting organometallic complexes for use in light emitting devices, in the name of S.Q. Shi issued January 23, 1996. **(Reference 25: Organometallic Oxinoids Patent [S4], col. 1, ll. 41-50)**.

For these reasons, both the JP-145 and the Prior Iridium OLED Patent independently destroy the novelty of claims 1 & 16. These claims are unpatentable.

Prior art showing OLEDs using *phosphorescent organometallic* osmium *compounds*

Before EP-238 was filed, Y. Ma, H. Zhang, J. Shen, C. Che, Electroluminescence from triplet metal-ligand charge-transfer excited state of transition metal complexes, Synthetic Metals 94:245-48 (1998) described using four *phosphorescent organometallic* **osmium** *compounds* in an OLED. **(Reference 23: Ma [D1]**, ***see also* Fig. 1 for the OLED structure**).



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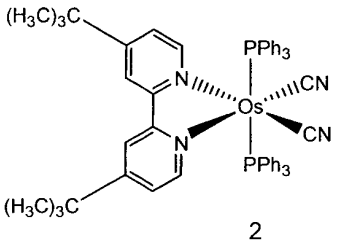

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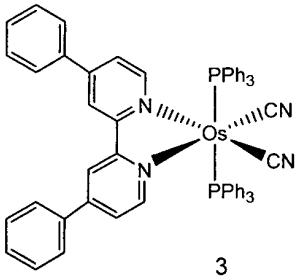

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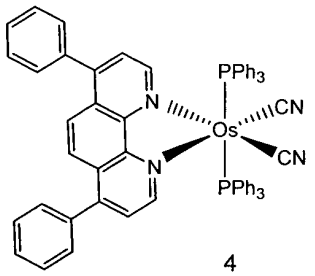

4

In particular, Ma fabricated an OLED having a heterostructure comprising, between an aluminum cathode and an ITO anode, two layers. The hole transport layer (HTL) comprises poly(N-vinyl carbazole) and the emissive *organometallic osmium compounds* (**1**)-(**4**). The electron transport layer (ETL) comprises 2-(4-biphenyl)-5-(4-tert-butyl-phenyl)-1,3,4-oxadiazole. Each osmium compound (**1**)-(**4**) contains an osmium atom (Os) bonded to at least three different organic ligands chosen from bipyridine and its derivatives (see compounds (**1**)-(**3**) leftmost group), 4,7-diphenyl-1,10-phenanthroline (see compound (**4**) leftmost group), triphenyl phosphine (see PPh_3 in compounds (**1**)-(**4**) at the 12 & 6 o'clock groups), and cyanide (see CN in compounds (**1**)-(**4**) 2 & 4 o'clock groups). Even under the classical definition of *organometallic*, CN is an organyl group bonded to a metal osmium (Os) thus making each of the compounds *organometallic osmium compounds*. Moreover, some co-inventors of EP-238 admitted that Ma's complexes are "organometallic." (**Reference 24: Thompson [D29], p. 173, Fig. 42**).

Ma determined that the osmium compounds are *phosphorescent*. In other words, Ma's emissive *organometallic osmium compounds* (**1**)-(**4**) are emissive *phosphorescent organometallic osmium compounds* per claim 16.

For these reasons, Ma destroys the novelty of claims 1 & 16. These claims are unpatentable.

Prior art renders non-inventive OLEDs using *phosphorescent organometallic* iridium-ppy *compounds* like $Ir(ppy)_3$

At the outset, it is worth noting that EP-238 is not a pioneering invention. All the component parts were known, and the inventors of EP-238 merely combined them. Ma, the closest prior art, sets forth the theory for *phosphorescent* OLED and exemplifies four OLEDs containing one of four different *phosphorescent organometallic* ***osmium*** *compounds*.

Ma teaches to make a simple substitution of any of its four *phosphorescent organometallic* **osmium** *compounds* (**1**)-(**4**) for a *phosphorescent organometallic* ***iridium*** *compound* of G. Di Marco, M. Lanza, and M. Pieruccini, A luminescent Iridium(III) cyclometallated Complex Immobilized in a Polymeric Matrix as a Solid-State Oxygen Sensor,

Adv. Mater. 8(7):576-79 (1996). (**Reference 27: DiMarco [D9]**). In particular, Ma states that the particular organometallic compounds of DiMarco "provide possibility to design the high-efficiency EL device by using ... triplet excited-state materials." (**Reference 23: Ma [D1], p. 245, col. 1**). Ma attributes the efficiency to the well-known use of a "strong interaction between metal centre and the ligands, [because] the transition metal complexes (such as Ru, Os, Ir) exhibit a metal-ligand charge-transfer (MLCT) excited state which shows the triplet nature." (**Reference 23: Ma [D1], p. 245, col. 1**). Ma claims to describe the "first observation of EL from triplet MLCT excited states of transition metal complexes." (**Reference 23: Ma [D1], p. 245, col. 2**).

DiMarco's *phosphorescent organometallic* ***iridium*** *compound* is $Ir(ppy)_2(dpt\text{-}NH_2)(PF_6)$. DiMarco's *phosphorescent organometallic* ***iridium*** *compound* has two ligands called ppy, the very same ppy used in EP-238's $Ir(ppy)_3$. Substituting DiMarco's *phosphorescent organometallic* ***iridium*** *compound* for one of Ma's *phosphorescent organometallic* **osmium** *compounds* would result in an OLED heterostructure of claim 16 in EP-238.

It would have been non-inventive to simply substitute one of Ma's exemplified osmium (II) complexes (**1**)-(**4**) with the iridium (III) complex of DiMarco in the hopes of finding an alternative to Ma's *phosphorescent organometallic* ***osmium*** *compound.*

Recall, the experimental basis for EP-238 includes the testing of one compound in multiple OLED architectures. If EP-238's experiment with a single iridium compound would have suggested (to them) to use any other *iridium* and *osmium* compound, then it seems perfectly natural for one skilled in the art having knowledge of Ma's OLED containing one of Ma's four osmium compounds to look to iridium compounds, especially when the one true pioneer, Ma, directed everyone to use a specific *organometallic phosphorescent iridium compound* comprising ppy.

EP-238's claims embrace a tremendous amount of subject matter, and the *iridium compounds* and *osmium compounds* are not confined to a structure, stability, luminescence, emission lifetime, or color of emission. The facts and law do not support a claim to such a broad scope of protection, especially since EP-238's contribution is a simple substitution of known components.

Moreover, DiMarco's *phosphorescent organometallic* ***iridium*** *compound* has 2 ppy ligands, while a narrower claim of the EP-238 recites $Ir(ppy)_3$ having three ligands ppy. This difference is meaningless. Any supplier of iridium could make and deliver an iridium compound having 2 or 3 ppy. See, e.g., A.P. Wilde, J. Phys. Chem. 95:629-34 (1991) (**Reference 30: Wilde**). In fact, EP-238 admitted that $Ir(ppy)_3$ was a known phosphorescent organometallic iridium compound. (**Reference 1: EP-238, ¶28**)(citing **Reference 45: King [D12]** to confirm that the phosphorescent organometallic iridium compound is indeed phosphorescent.) The idea of adding another ppy is not grounds of any claim of invention of an iridium emitter that is different than DiMarco and much less ownership of all iridium compounds in all meaningful OLED.

Anyone reading Ma and DiMarco could make the simple substitution proposed by Ma and result in what EP-238 claimed. Simple substitutions that are directed by previous research with expected results are not inventive.

Conclusion

For the reasons stated above, the EP-238 patent should be completely revoked.

Respectfully submitted,

LOWE HAUPTMAN & HAM, LLP

Sean A. Passino

Randy A. Noranbrock

SAP/RAN:drs
Enclosures: Third Party Observations;
references 5-28, 30-31, & 42-45