

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**  
**BEFORE THE PATENT TRIAL AND APPEAL BOARD**

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In re *Inter Partes* Review of:

U.S. Patent No. 8,558,223

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**REPLY DECLARATION OF BARRY P. RAND, PH.D.**

***PROTECTIVE ORDER MATERIAL***

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U.S. Patent No. 8,558,223  
Declaration of Dr. Barry P. Rand

## **I. INTRODUCTION**

I, Barry P. Rand, Ph.D., declare as follows:

1. I have been asked to provide this reply declaration in order to address specific issues raised by Patent Owner (“PO”) and its witness Dr. Schanze relating to the Werner and Ma references that were addressed in my Opening Declaration (Ex.1003).

## **II. BACKGROUND AND QUALIFICATIONS**

2. My background and qualifications are detailed in my Opening Declaration.

## **III. RELEVANT LEGAL STANDARDS**

3. I am not an attorney. I have been informed about certain aspects of patent law, which have been discussed in my Opening Declaration (Ex.1003). My understanding of the law remains the same and I have continued to apply this understanding of the law for purposes of this reply declaration.

## **IV. LEVEL OF ORDINARY SKILL IN THE ART**

4. I applied the same definition of a person of ordinary skill in the art (“POSA”) as explained in my Opening Declaration (Ex.1003, ¶46) for purposes of this reply declaration.

**V. WERNER**

**A. Werner directly teaches or at least motivates with a reasonable expectation of success using the metal complex  $W_2(hpp)_4$  for doping the organic compound BPhen**

**1. Werner discloses metal complexes and organic compounds that satisfy the Challenged Claims**

5. As I discussed in my Opening Declaration,  $W_2(hpp)_4$  and BPhen disclosed in Werner satisfy the Challenged Claims. *See generally*, Ex.1003, Ground I; *see, e.g., id.*, ¶¶66–73 (discussing why  $W_2(hpp)_4$  and BPhen satisfy claim 1). I understand that Pictiva primarily responded that despite Werner’s listing of  $W_2(hpp)_4$  and BPhen as “candidate” materials, Werner does not specifically link these two materials. POR,<sup>1</sup> 2. It is my opinion that in so doing, Pictiva ignores the wording of the claims (Claim 1, for example, requires no specific metal complex or organic compound) and the broader teachings in Werner.  $W_2(hpp)_4$  and BPhen are not the only compounds that satisfy the Challenged Claims, nor are they called out in the Challenged Claims, except for claim 6 (which, however, can be also satisfied by 12 other specific compounds). Ex.1021 [Schanze Tr.], 17:5–18:9, 25:3–26:11, 27:8–30:5.

6. For example, it is my opinion that all the metal complexes disclosed in Werner satisfy claim 1. Claim 1 only requires a metal complex with a central metal atom. Werner discloses in the Abstract that “a neutral electron-rich metal complex having a central atom” is suitable for its invention, and all of the metal complexes in

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<sup>1</sup> Patent Owner Response (Paper 19).

Werner satisfy that. Ex.1005 [Werner], Abstract. Specifically, Werner discloses 8 specific classes of metal complexes: pi-complexes with a central metal atom (Ex.1005, 7:29–11:4), paddlewheel complexes and multinuclear complexes in general (*id.*, 11:6–19:29), metal complexes with a central metal atom and 6 different classes of ligands (*id.*, 19:33–20:14), such as carboranes (*id.*, 20:22–26), borates (*id.*, 20:28–23:22), tripod ligands (*id.*, 24:1–25:16), triazacycloalkanes (*id.*, 25:18–26:14), fullerenes (*id.*, 26:16–24), and pincer ligands (*id.*, 26:26–36:17). All of these metal complexes have a central metal atom (*id.*, 20:16–20).

7. Further, Werner specifically discloses exemplary “paddlewheel complexes,” such as “M<sub>2</sub>hpp<sub>4</sub> with M equal to Cr, Mn, Fe, Co, Ni, Cu, Zn, Mo, or W,” as suitable metal complexes. Ex.1005, 60:20–21. When M is Cr, Mo, or W, the metal complex has two central metal atoms selected from Cr, Mo, or W, which satisfies claim 4.

8. It is my opinion that as for the matrix materials, at least “aryl-substituted, such as phenyl or naphthyl-substituted” phenanthrolines, such as BPhen, and “aryl-substituted, such as phenyl or naphthyl-substituted” heteroaromatics, such as TAZ (*id.*, 54 (figure); 55:3–17), that Werner discloses, are organic compounds comprising a heteroaromatic compound which is conjugated with an aromatic compound via a C—C bond, which satisfy claim 1. I understand that Dr. Schanze also agreed on this point. *See, e.g.*, Ex.1021, 93:16–94:7 (“If [you] consider the

structure, phenanthroline – 110 phenanthroline as being a heteroaromatic compound taken as a whole, then substitution at the five or seven positions with a phenyl substituent could be considered to be a heteroaromatic compound conjugated to an aromatic compound via a CC bond.”); *id.*, 96:13–97:4 (“[I]f it’s a -- structure is a heteroaromatic compound, that’s substituted at a position other than the nitrogen with a aromatic ring such as phenyl, that would constitute the definition of a heteroaromatic compound that’s substituted with an aromatic compound, or linked to, via a CC bond.”).

9. Claim 6 requires that the organic compound is selected from among 13 compounds, of which BPhen is compound VI (Ex.1003, ¶151); and aryl- or naphthyl-substituted phenanthroline derivatives are compounds V, VI and XI.

**2. The prior art demonstrates that W<sub>2</sub>(hpp)<sub>4</sub> increased conductivity when combined with BPhen or other phenanthroline derivatives.**

10. I understand that Pictiva argues that a POSA would not have combined W<sub>2</sub>(hpp)<sub>4</sub> and BPhen, based on the disclosures in two Novald patents, U.S. 7,911,129 (“’129 patent”; Ex.2043) and U.S. 7,972,541 (“’541 patent”; Ex.2044).<sup>2</sup> These patents do not support Pictiva’s position.

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<sup>2</sup> I have also reviewed Ex.1035, which is the English translation of WO2007054345A, which I understand is the application that lead to Ex.2044 and was published on May 18, 2007. In pertinent part, the translation is essentially verbatim to the text I reviewed in Ex.2044.

11. The '541 patent discloses that the conductivity of W<sub>2</sub>(hpp)<sub>4</sub>-doped BPhen was significantly higher than undoped BPhen. Ex.2044, 4:20–43 (Table 1), 10:6–19. Specifically, the conductivity of undoped BPhen was less than 1 x 1<sup>-10</sup> S/cm. Ex.2044, 4:20–43, Table 1 (reproduced below). The '541 patent reports two different results for conductivity of W<sub>2</sub>(hpp)<sub>4</sub>-doped BPhen: Table 1 discloses that conductivity of an OLED with 5 mol% doping of W<sub>2</sub>(hpp)<sub>4</sub> in BPhen was 4 x 10<sup>-9</sup> S/cm, which is at least a 40-fold increase in conductivity compared to undoped BPhen.

TABLE 1

| CV data, empirically determined LUMO energies and measured conductivities of various electron transport materials (BAIq <sub>2</sub> = bis(2-methyl-8-quinolinato)-4-(phenylphenolate) aluminium (III), BPhen = bathophenanthroline, Alq <sub>3</sub> = tris (8-hydroxyquinoline) aluminium, Cl6-Alq = tris (5,7-dichloro-8-hydroxyquinoline) aluminium) |                                                                                     |                                     |                                                                          |
|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------|-------------------------------------|--------------------------------------------------------------------------|
| Matrix material                                                                                                                                                                                                                                                                                                                                          | LUMO in eV<br>(determined by CV<br>with Fe/Fe <sup>+</sup> as<br>internal standard) | σ (conductivity)<br>in S/cm undoped | σ (conductivity) in<br>S/cm with a doping<br>concentration of<br>5 mol % |
| Alq <sub>3</sub>                                                                                                                                                                                                                                                                                                                                         | 2.4                                                                                 | <1E-10                              | 9.2E-8                                                                   |
| BPhen                                                                                                                                                                                                                                                                                                                                                    | 2.42                                                                                | <1E-10                              | 4E-9                                                                     |
| BAIq <sub>2</sub>                                                                                                                                                                                                                                                                                                                                        | 2.39                                                                                | <1E-10                              | 8e-8                                                                     |

Ex.2044, 4:20–43 (annotated). Comparative Example 2 (Ex.2044, 10:6–19) discloses that when an OLED was made “in a manner analogous ... to Example 2 [*id.*, 9:55–10:5], but BPhen was used instead of the matrix material Ie,” (i.e., 4 mol%

doping of  $W_2(hpp)_4$  in BPhen), the conductivity was  $4 \times 10^{-7}$  S/cm, which was at least a 4000-fold increase in conductivity compared to undoped BPhen ( $<1 \times 10^{-10}$ , Ex.2044, Table 1). Thus, the '541 patent demonstrates that researchers observed before the '223 patent that merely 4 mol% doping of  $W_2(hpp)_4$  in BPhen increased conductivity of an electron conducting layer by at least 4000-fold. It is my opinion that a POSA would understand the 4000-fold increase as a significant improvement in conductivity.

12. By way of comparison, Ma measured an increase in conductivity<sup>3</sup> by changing the percentage of a dopant (Liq) from 0% (i.e., undoped BPhen) to 50 wt% (about 68 mol%), in BPhen. Ex.1007, 812–813, Fig. 5. The result shows that the maximum increase in current density was observed when 33 wt% (about 50 mol%) Liq was doped in BPhen, which was about a two to three-fold increase (e.g., from  $\sim 130$  mA/cm<sup>2</sup> to  $\sim 370$  mA/cm<sup>2</sup> at 12 V) over undoped (pristine) BPhen. *Id.* Ma summarizes that “the device with the doping ratio of 33 wt% Liq demonstrates the best current density vs voltage (J–V) curve among all the electron only devices,” and concluded that it was “a significant improvement in the device performance as

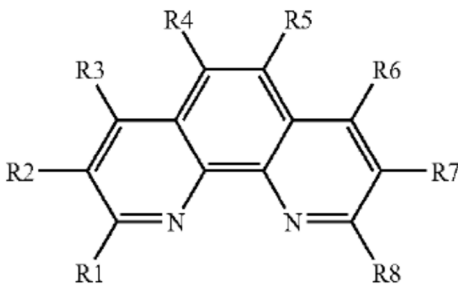
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<sup>3</sup> Ma Figure 5 reports current density as a function of voltage. Current density (mA/cm<sup>2</sup>) and conductivity ( $\sigma$ ) are directly related through a variation of Ohm's law where  $J = \sigma E$  (where J is current density,  $\sigma$  is conductivity and E is the electric field). Accordingly, a two- to three-fold increase in current density would result in a two- to three-fold fold increase in conductivity, and thus it is acceptable to compare current density increases in Ma with conductivity increases in the '541 patent.

compared with the referenced device.” *Id.*, 810–811. In other words, researchers would consider about a two to three-fold increase in conductivity as a significant improvement. Compared to this result, a POSA would consider that the 4000-fold increase in conductivity obtained in the ’541 patent by  $W_2(hpp)_4$ -doped BPhen was a significant improvement. Further, the 4000-fold increase was obtained by merely 4 mol% doping of  $W_2(hpp)_4$ , compared to 50 mol% doping of Liq in Ma. Thus, a POSA would conclude that  $W_2(hpp)_4$  is a significantly more efficient dopant than Liq for BPhen.

13. Further, the ’541 patent discloses that when “[s]elective phenanthroline derivatives” were used as matrix materials for doping with 5%  $W_2(hpp)_4$ , a “considerably improved conductivity value[]” with each of the phenanthroline derivatives was observed, as shown in Table 2 below:

TABLE 2

| structure I                                                                       |                          |                          |              |                         |  |
|-----------------------------------------------------------------------------------|--------------------------|--------------------------|--------------|-------------------------|--|
|  |                          |                          |              |                         |  |
| R                                                                                 | T <sub>g</sub><br>(° C.) | T <sub>s</sub><br>(° C.) | LUMO<br>(eV) | Conductivity<br>(S/cm)* |  |
| Ia R2, R4, R5, R7 = H<br>R1, R3, R6, R8 = phenyl                                  | 106                      | 260                      | 2.47         | 3E-4                    |  |
| Ib R2, R4, R5, R7, R8 = H<br>R3, R6 = phenyl<br>R1 = 1-naphthyl                   | 104                      | 265                      | 2.42         | 5E-6                    |  |
| Ic R2, R4, R5, R7 = H<br>R3, R6 = phenyl<br>R1, R8 = naphthyl                     | 119                      | 280                      | 2.5          | 2E-6                    |  |
| Id R2, R4, R5, R7 = H<br>R3, R6 = phenyl<br>R1, R8 = 1-(4,4'-biphenyl)            | 140                      | 317                      | 2.53         | 6E-5                    |  |
| Ie R2, R4, R5, R7 = H<br>R3, R6 = phenyl<br>R1, R8 = 1-phenanthryl                | 165                      | 317                      | 2.51         | 2E-6                    |  |

\*Concentration IVa (W<sub>2</sub>(hpp)<sub>4</sub>): 5 mol %

Ex.2044, 9:16–54, Table 2. All of the phenanthroline derivatives listed in Table 2 are aryl-substituted phenanthrolines (particularly phenyl- and naphthyl-substituted phenanthrolines), which Werner (Ex.1005) discloses as suitable matrix materials for its invention, and are “organic compounds compris[ing] a heteroaromatic compound which is conjugated with an aromatic compound via a C—C bond,” which satisfy claim 1. Ex.1005, 55:3–6; Ex.1001, 19:20–23, claim 1. The '541 patent (Ex.2044) demonstrates that phenanthroline films (one of the disclosed matrix material classes in Werner) other than BPhen, when doped with W<sub>2</sub>(hpp)<sub>4</sub> also showed significant

conductivity, further confirming that prior to the '223 patent, a POSA would expect that  $W_2(hpp)_4$ -doped phenanthrolines (including BPhen) would have a high degree of electron conductivity, as Werner also teaches. Ex.1005, 55:3–6.

14. Further, the '541 patent compares one of its preferred matrix materials,  $W_2(hpp)_4$ -doped *Ie* (from Table 2 above) with  $W_2(hpp)_4$ -doped BPhen in Comparative Example 2. Ex.2044, 9:55–10:19. As discussed above, the conductivity of  $W_2(hpp)_4$ -doped BPhen was  $4 \times 10^{-7}$  S/cm (Ex.2044, 10:6–12), and this value is at least 4000-fold higher than the conductivity of the undoped BPhen (less than  $1 \times 10^{-10}$  S/cm) (Ex.2044, 4:40, Table 1).<sup>4</sup> The inventors of the '541 patent observed that “[w]hen using the matrix material *Ie*, the conductivity is increased by a factor of 5,” which they deemed made it of sufficient conductivity for use in an OLED. Ex.2044, 10:17–19, 8:62-67, *see also* claim 29 (claiming an OLED using a semiconductor material according to claim 1, which covers use of *Ie*). These results show that  $W_2(hpp)_4$ -doping resulted in a more significant increase in conductivity (3 orders of magnitude increase (4000x) compared to undoped BPhen) than changing the matrix material while keeping the dopant constant (less than an order of magnitude increase (5x) when the doping material and concentration was the same across two different

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<sup>4</sup> To be sure, I understand that the inventors concluded that “the conductivity of a semiconductor material consisting of  $[W_2(hpp)_4]:BPhen$  is too low for use in organic light-emitting diodes.” Ex.2044, 10:15–17. As a practical matter, I do not agree with the inventor’s assessment, as a 4000x increase in conductivity is quite significant, and would be expected to be effective for use in an OLED.

matrix materials). Thus, it is my opinion that a POSA would have concluded based on the results disclosed in the '541 patent that  $W_2(hpp)_4$  is a suitable dopant for BPhen as well as for other phenanthroline derivatives (i.e., heteroaromatic compounds conjugated with an aromatic compound via a C—C bond).

15. As I discussed in my Opening Declaration, given the structural similarities between Compound V (BCP) and Compound VI (BPhen), the charge transfer reported in the '223 Patent between  $W_2(hpp)_4$  and BCP provides further confirmation of my opinion that a POSA would reasonably expect charge transfer between  $W_2(hpp)_4$  and BPhen. Ex.1003, ¶¶114–115 (citing Ex.1001, 16:56–17:21). BCP is also a phenyl-substituted phenanthroline that Werner discloses as a suitable matrix material. Ex.1005, 55:3–6. As Dr. Schanze testified, BCP was used for semiconducting layers as of 2005. Ex.1021, 31:1–3 (“Q. And BCP was a compound that was known for use in semiconducting layers as of 2005? A. Yes, Ma’am. I would agree with that.”). It was well known that BCP and BPhen have almost the same HOMO and LUMO levels and comparable electron-transport properties. Ex.1033, 164704–6 (“comparable electron-transport properties can be expected in BCP and BPhen.”); Ex.1034, 094506-1 (“BPhen is selected because its highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels are almost the same as those of BCP.”).

16. Therefore, the foregoing materials further support my opinion that a POSA would expect that BPhen would be a suitable matrix material for  $W_2(hpp)_4$ .

**3. The prior art does not teach away from combining  $W_2(hpp)_4$  and BPhen**

17. I understand that Pictiva asserts that the POSA would not have considered  $W_2(hpp)_4$  to be an “effective” n-dopant for BPhen in OLEDs. POR, 10–15. I disagree.

**a)  $W_2(hpp)_4$  does not destroy organic compounds in electronics**

18. I understand that Pictiva argues that “ $W_2(hpp)_4$  was also known to destroy organic compounds” based on its reaction with  $CH_2Cl_2$  or  $C_{60}$  in solution. POR, 13 (citing Ex.2008). In my opinion, interactions between  $W_2(hpp)_4$  and  $CH_2Cl_2$  or  $C_{60}$  in solution would have no meaningful impact on interactions between  $W_2(hpp)_4$  and BPhen in the gas-phase when they are jointly vaporized. Further, I understand that Dr. Schanze testified that  $CH_2Cl_2$  is not commonly used for making a doped ETL. Ex.1021, 147:12–14 (“Q. Is using methylene chloride commonly used for making a doped ETL? A. To my knowledge, no. It’s not.”).

19. Additionally, while Pictiva argues that  $W_2(hpp)_4$  “reduces  $C_{60}$  to intractable reduction products” (POR, 13), Pictiva’s own evidence demonstrates that  $C_{60}$  doped by  $W_2(hpp)_4$  can yield an effective charge transfer. For example, as Dr. Schanze admits, the “abstract of Reference 121 [Ex.2016] indicates that authors studied  $C_{60}$  layer doped with  $W_2(hpp)_4$ .” Ex.2041, ¶82 (citing Ex.2016). Menke 2012

(Ex.1022) (which is Ref. 122 of Ex.2003) reports that “[v]ery high conductivities of up to 4 S/cm are achieved for both  $[\text{Cr}_2(\text{hpp})_4]$  and  $\text{W}_2(\text{hpp})_4$ ” as n-dopants for  $\text{C}_{60}$ . Ex.1022, Abstract. Moreover, Pictiva’s assertion is contradicted by Dr. Schanze’s argument that “research groups also studied doping of  $\text{C}_{60}$  with  $\text{W}_2(\text{hpp})_4$ ,” because “ $\text{C}_{60}$ ’s ... energy [level is] below the ionization energy of  $\text{W}_2(\text{hpp})_4$ , making the electron transfer from  $\text{W}_2(\text{hpp})_4$  to  $[\text{C}_{60}]$  energetically favorable.” Ex.2041, ¶¶81–82 (citing to ref. 121 [Ex.2016], ref. 122 [Ex.1022], and ref. 126 [Ex.2009] of Ex.2003). If  $\text{W}_2(\text{hpp})_4$  reduced  $\text{C}_{60}$ , other groups would not be studying doping it with  $\text{W}_2(\text{hpp})_4$ .

**b) The HOMO of  $\text{W}_2(\text{hpp})_4$  and LUMO of BPhen are close enough to permit electron transfer**

20. I understand that Pictiva also argues that “to effect n-doping, the ionization potential (“IP”) or Highest-Occupied-Molecular-Orbital (HOMO) energy level of  $\text{W}_2(\text{hpp})_4$  should be no more than 0.5 eV lower than the electron affinity (“EA”) or Lowest-Unoccupied-Molecular-Energy (LUMO) of BPhen/TAZ.” Pictiva further argues that “the electron affinities of BPhen and TAZ are less than the ionization potential of  $\text{W}_2(\text{hpp})_4$  (3.76 eV) minus 0.5 eV, not meeting ‘the requirement of the doping capability’ prescribed by Mr. Werner himself,” based on the referenced Novaled patents, U.S. 7,911,129 (“129 patent”) and U.S. 7,972,541 (“541 patent”). POR at 3, 11; Ex.2041, ¶¶20–25. I disagree for the following reasons.

21. *First*, Dr. Schanze used different HOMO values for  $\text{W}_2(\text{hpp})_4$  between his declaration for POPR (Ex.2001) and the one for POR (Ex.2041). In his

declaration for POR, Dr. Schanze argued that “W<sub>2</sub>(hpp)<sub>4</sub> was reported to have an onset of ionization in the gas phase at about 3.51 eV and a peak gas phase ionization energy of about 3.76 eVs” and “[t]hus, its HOMO would be considered to be have an energy level about 3.76 eV.” Ex.2041, ¶¶ 66–67 (citing to Ex.2004, 770). In other words, Dr. Schanze selected the “peak gas phase ionization energy” value for the HOMO in his second declaration, as he confirmed. Ex.1021, 108:10–13 (“Q. Right. So my question is, 3.76eV is the HOMO level that you have used in the course of developing your opinions. Right? A. That’s correct.).

22. This contradicts his prior statement in his declaration for the POPR, where he asserted that

[t]he HOMO level is the energy level of the highest occupied molecular orbital, and according to Koopman’s theorem, the first ionization energy of a molecule is equal to the negative of the energy of the HOMO of the molecule. So for W<sub>2</sub>(hpp)<sub>4</sub>, because it was reported to have an *onset of ionization at about 3.51eV* (see EX2004-Cotton-teaxtbook, 189), its *HOMO would be considered to have an energy level about 3.51eV*.<sup>5</sup>

Ex.2001, ¶47 (emphasis added). *Compare* image at Ex.2001 ¶47 *with* image at Ex.2041 ¶67 (reporting different HOMO levels for W<sub>2</sub>(hpp)<sub>4</sub>). In both of his declarations, although noting that there is reported variation in the LUMO levels

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<sup>5</sup> Out of the two values cited by Dr. Schanze, I agree that the onset of ionization value of 3.51 eV more appropriately reflects the HOMO of W<sub>2</sub>(hpp)<sub>4</sub>.

reported for BPhen, he used 3.0 eV as the LUMO value for BPhen. Ex.2001, ¶¶52, 54, Ex.2041, ¶¶23, 60, 63–67.

23. This change is meaningful in the context of Pictiva's argument. I observe that the delta between HOMO:LUMO when the  $W_2(hpp)_4$  is 3.51 eV (as Dr. Schanze used in his first declaration) and BPhen is 3.0 eV is 0.51 eV, which is about 0.5 eV, and thus effectively the same as the limit Pictiva argues is required to effect electron transfer. In contrast, when Dr. Schanze used the 3.76 eV value for  $W_2(hpp)_4$  in his second declaration, the delta between  $W_2(hpp)_4$  and BPhen is 0.76, which is outside of the limit Pictiva proposes is appropriate for an electron conducting layer.

24. *Second*, the '541 patent shows that either the difference between the HOMO and LUMO levels of a dopant and a matrix material does not need to be less than 0.5 eV to achieve electron transfer, or the difference between the HOMO of  $W_2(hpp)_4$  and the LUMO of BPhen is within the 0.5 eV difference; in any event, the data in the '541 patent confirms for the POSA that a  $W_2(hpp)_4$ :BPhen film would be expected to conduct electrons.

25. The '541 patent (Ex.2044), cited by Dr. Schanze, repeats Pictiva's assertions (as cited from Ex.2043) regarding the purported need for a no-more-than-0.5 eV difference between the HOMO:LUMO levels of dopant and matrix material to achieve electron transfer. Ex.2041, ¶20 (citing Ex.2043, 10:56–63); *id.*, ¶22 (citing Ex.2044, 8:19–23).

26. The '541 patent explains that each of the disclosed phenanthrolines' LUMO "falls within the range which means it can be doped with ... in particular [W<sub>2</sub>(hpp)<sub>4</sub>]" (Ex.2044, 8:46–53), and discloses in Table 2 exemplary phenanthroline derivatives, which "show[ed] considerably improved conductivity values ... according to the invention." Ex.2044, 9:16–53, Table 2 (reproduced below).

TABLE 2

structure I

| R                                                                      | T <sub>g</sub><br>(° C.) | T <sub>s</sub><br>(° C.) | LUMO<br>(eV) | Conductivity<br>(S/cm)* |
|------------------------------------------------------------------------|--------------------------|--------------------------|--------------|-------------------------|
| Ia R2, R4, R5, R7 = H<br>R1, R3, R6, R8 = phenyl                       | 106                      | 260                      | 2.47         | 3E-4                    |
| Ib R2, R4, R5, R7, R8 = H<br>R3, R6 = phenyl<br>R1 = 1-naphthyl        | 104                      | 265                      | 2.42         | 5E-6                    |
| Ic R2, R4, R5, R7 = H<br>R3, R6 = phenyl<br>R1, R8 = naphthyl          | 119                      | 280                      | 2.5          | 2E-6                    |
| Id R2, R4, R5, R7 = H<br>R3, R6 = phenyl<br>R1, R8 = 1-(4,4'-biphenyl) | 140                      | 317                      | 2.53         | 6E-5                    |
| Ie R2, R4, R5, R7 = H<br>R3, R6 = phenyl<br>R1, R8 = 1-phenanthryl     | 165                      | 317                      | 2.51         | 2E-6                    |

\*Concentration IVa (W<sub>2</sub>(hnp)<sub>4</sub>): 5 mol %

\*Concentration IVa (W<sub>2</sub>(hpp)<sub>4</sub>): 5 mol %

Ex.2044, 9:22–49, Table 2. As can be seen in Table 2, the LUMO values of the exemplary matrix materials are between 2.42 eV to 2.53 eV. Table 1 of the '541 patent discloses that the LUMO value of BPhen is comparable to the phenanthrolines in Table 2, with a LUMO of 2.42 eV.

27. The '541 patent discloses that the gas phase ionization potential of  $W_2(hpp)_4$  is 3.6 eV while the solid-phase ionization potential is estimated to be approximately 2.5 eV. Ex.2044, 4:15–19. Thus, the difference between these two HOMO values is 1.1 eV. This is a particularly significant difference if the criteria for expected electron conductivity is whether the difference between HOMO and LUMO is bigger than 0.5 eV or not. The following table summarizes the LUMO values of BPhen and the preferred compounds Ia to Ie in the '541 patent and the difference between the HOMO of  $W_2(hpp)_4$  and the LUMO of each matrix material using the two different HOMO values of  $W_2(hpp)_4$ .

| Matrix material | LUMO value | The difference between HOMO and LUMO |                                |
|-----------------|------------|--------------------------------------|--------------------------------|
|                 |            | if $W_2(hpp)_4$ HOMO is 3.6 eV       | if $W_2(hpp)_4$ HOMO is 2.5 eV |
| BPhen           | 2.42       | 1.18                                 | 0.08                           |
| Ia              | 2.47       | 1.13                                 | 0.03                           |
| Ib              | 2.42       | 1.18                                 | 0.08                           |
| Ic              | 2.5        | 1.1                                  | 0                              |
| Id              | 2.53       | 1.07                                 | 0.03                           |
| Ie              | 2.51       | 1.09                                 | 0.01                           |

Ex.2044, 4:40 (Table 1), 9:24–49 (Table 2). Thus, if the HOMO of  $W_2(hpp)_4$  is 3.6 eV, all of these phenanthroline compounds have the HOMO-LUMO difference of

**more than 0.5 eV.**<sup>6</sup> Under Pictiva's argument that the delta between the HOMO and LUMO cannot be more than 0.5 eV, there should be no electron transfer – except the data from the '541 patent shows that there is improved conductivity for each of these  $W_2(hpp)_4$ -doped phenanthrolines. Ex.2044 at Table 1, Table 2. If the HOMO of  $W_2(hpp)_4$  is 2.5 eV, all of these phenanthrolines have the HOMO-LUMO difference of **less than 0.5 eV**. Under Pictiva's argument that the delta between the HOMO and LUMO cannot be more than 0.5 eV to permit electron transfer, there should be good electron transfer for each of these  $W_2(hpp)_4$ -doped phenanthrolines – which is exactly what Ex.2044 shows (Table 1, Table 2).

28. This suggests that that the HOMO of  $W_2(hpp)_4$  is 2.5 eV and the LUMO of the phenanthrolines (including BPhen) is around 2.4–2.55 eV.

29. In any event, the '541 patent tells the POSA that the  $W_2(hpp)_4$  doped phenanthroline derivative films in Table 2 showed “considerably improved conductivity values,” meaning these films transferred electrons. Ex.2044, 9:50–53. Moreover, as discussed above in **Section V.A.2**, the '541 patent demonstrates that  $W_2(hpp)_4$ -doped BPhen showed a 4000-fold increase in conductivity compared to undoped BPhen, which a POSA would consider a significant improvement. This confirms that whatever the difference is between the HOMO of  $W_2(hpp)_4$  and the

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<sup>6</sup> I note that these values would also contradict the statement in the '541 Patent that each of the disclosed phenanthrolines' LUMO “falls within the range which means it can be doped with ... in particular [ $W_2(hpp)_4$ ]” (Ex.2044, 8:46–53).

LUMO of BPhen (or the other phenanthrolines), it is sufficient to permit electron transfer. Thus, even if the “proper” HOMO value of  $W_2(hpp)_4$  is above 3.5 eV, and the “proper” LUMO value of BPhen is below 3.0 eV, a  $W_2(hpp)_4$ :BPhen film will conduct electrons. Therefore, a POSA would not be dissuaded by the HOMO and LUMO values for  $W_2(hpp)_4$  and BPhen for use of both together in an ECL.

30. Indeed, Dr. Schanze admitted that the HOMO-LUMO relationship is not always important in electron transfer for ECL and there are other mechanisms for electron transfer, such as LUMO-LUMO transfer. For example, in reference to Ma (Ex.1007) which used Liq as an n-dopant for BPhen in ECL, Dr. Schanze testified that he did not check the HOMO level of Liq because it is not relevant.

Q. Okay. Why didn't you look up the HOMO of LiQ?

A. I didn't see how it was relevant.

Q. And why didn't you think it was relevant?

...

A. Well, in Ma -- this paper that we are referring to -- they attribute the differences to LUMO-LUMO interactions. And if you read down in that paragraph, they state that:

“In a single host device, electrons hop along the LUMO and BPhen, since the LUMO-LUMO difference between BPhen and LiQ is neglectable, subject to their similar LUMO, transport manifolds along with their LUMO are expected to exhibit a certain extent of overlap, after a mixing ratio.”

So they are attributing the effect to a LUMO-LUMO interaction.

So in that case, the HOMO is not relevant.

Ex.1021, 58:25–59:18. However, Pictiva fails to explain why it assumed the HOMO-LUMO transfer is the only possible electron transfer mechanism in  $W_2(hpp)_4$  and BPhen.

**B. Joint vaporization of  $W_2(hpp)_4$  and BPhen**

31. I understand that Pictiva argues that “[r]eported reaction conditions for  $W_2(hpp)_4$  [in Werner] also do not suggest use of joint vaporization.” POR, 16–17. I disagree. As I discussed in my Opening Declaration, Werner discloses that a metal complex and a matrix material suitable for its invention can be combined using co-evaporation (i.e., joint evaporation) and this disclosure is universal to the disclosed metal complexes and organic compounds, including  $W_2(hpp)_4$  and BPhen. Ex.1003, ¶70; Ex.1005, 56:12–18 (“[c]o-evaporation in vacuum with a source for the matrix material and one for the dopant”); 61:10–11 (“The provided metal complex is evaporated simultaneously with the matrix material”), 61:35–62:3 (“The neutral complex was sealed under protective gas in ampoules. Under exclusion of oxygen, this material was used to fill an evaporator source. Doped layers with a doping ratio of dopant:matrix material of 1:10 were produced by co-evaporation of matrix and dopant with ZnPc as the matrix material.”), 62:13–16 (“A layer of zinc-phthalocyanine (ZnPc) was doped with the metal complex  $[Cr_2(hpp)_4]$ . The doped layer was produced by co-evaporation of the ZnPc matrix and the dopant  $Cr_2(hpp)_4$  in high vacuum.”).

32. Further, Werner differentiates “co-evaporation” from “sequential evaporation” by teaching them separately. Ex.1005, 56:16–22 (“a) Co-evaporation in vacuum with a source for the matrix material and one for the dopant, b) Sequential deposition of the matrix material and the n-dopant on a substrate with subsequent diffusion of the dopant, in particular through thermal treatment.”). Thus, a POSA would *not* interpret that “co-evaporation” in Werner can be sequential evaporation.

33. Moreover, Pictiva concedes that Werner discloses that “[t]he provided metal complex is evaporated simultaneously with the matrix material,” POR, 15 (Ex.1005, 61:10–11), but fails to explain why simultaneous evaporation is not joint evaporation. Dr. Schanze has explained that when the materials are “deposited in sequence,” they are *not* “simultaneously” deposited. Ex.2041, ¶96 (arguing “fullerene and cobaltocene were deposited in sequence, not simultaneously, following method (b) of ‘Implementation of doping,’” referring to Example 3 in Ex.1005, 62:23–31, where a layer of fullerene was deposited first and then followed by a layer of cobaltocene). Thus, Dr. Schanze admitted that the simultaneous evaporation in Werner is not sequential vaporization, but joint vaporization.

34. I understand that Dr. Schanze has argued that, as of the priority date, the only use of  $W_2(hpp)_4$  was in solution, attempting to use this purported fact as evidence that  $W_2(hpp)_4$  and BPhen could not have been jointly vaporized. Ex.2041, ¶99. That is incorrect. As Pictiva admits, Cotton 2003 relates to chemistry in living organisms, not

electronics. POR, 16; Ex.2008 at 125 (“It should be pointed out that there might be some relationship of this work and chemistry in living organisms”). Thus, Ex.2008 (Cotton 2003) does not teach away from a gas-phase reaction with  $W_2(hpp)_4$  for making an organic electronic component.

35. Further, Ex.2008, Cotton 2003 does not conclude that  $W_2(hpp)_4$  *should* be used *only* in a solution. Rather, the authors recognized that using  $W_2(hpp)_4$  in organic solvents has a “potential” and “is an area of great current interest,” which clearly indicates that the use of  $W_2(hpp)_4$  in a solution was not established as of 2003. Ex.2008 at 124. In fact, Cotton et al. published an article in which a gas-phase reaction was performed with  $W_2(hpp)_4$ . Ex.2009, Ref. 16 (“Samples were handled and spectra were obtained using rigorous air-sensitive techniques . . .  $W_2(hpp)_4$  sublimed at 250° to 280°C.”). And, Ex.2044 describes using  $W_2(hpp)_4$  in particular with matrix materials having evaporation temperatures over 200 °C, (Ex.2044, 8:39-40) which suggests its use in the gas phase.

36. To the extent there is any question whether Werner’s disclosures are sufficient to demonstrate joint vaporization, it is my opinion that the POSA would have been motivated to jointly vaporize the metal complex and organic compound with a reasonable expectation of success. “Joint evaporation” is the most natural and straightforward interpretation of “[c]o-evaporization” (Ex.1005, 56:12–18) or “evaporated simultaneously” (*id.*, 61:10–11) for forming a layer, particularly when

there is nothing to indicate that materials were not jointly vaporized. Indeed, Dr. Schanze admitted in his report for a German proceeding<sup>7</sup> involving the German patent equivalent to the '223 patent,<sup>8</sup> that his use of “co-evaporation” would be understood to refer to joint vaporization, acknowledging that one “would not be able to determine” that he used joint evaporation from the description and that “*if one did such a ... sequential deposition, ... you would expect it to be stated.*” Ex.1021, 38:12–39:24 (emphasis added); *see also id.*, 36:10–21 (Dr. Schanze referring to his joint vaporization experiments as “co-evaporation,” e.g., “The experiments involved the co-evaporation of LiQ and the structure on the left, figure 2, which you are referring to as BTB”; “the aim of the experiment was to determine whether a coordination complex forms when LiQ and 742 and co-evaporated. Excuse me, when LiQ and compound BTB are co-evaporated.”), *see also id.* 38:12-15 (Q. “And you, as the person who oversaw this experiment, did you jointly -- jointly vaporize the B[T]B and the LiQ? A. To the best of my understanding, yes.”)

**C. Conductivity obtainable by n-doping BPhen with W<sub>2</sub>(hpp)<sub>4</sub>**

37. I understand that Pictiva contests whether Werner discloses the electrical conductivity of the ECL to the electrodes. POR, 17. It is my opinion that it does. Werner discloses “an organic light-emitting diode” using the disclosed ECLs, which

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<sup>7</sup> Filed in Munich Regional Court, Ref. 21 O2163/24 (hereinafter, “the German proceeding”); *see* paragraph 52, below.

<sup>8</sup> DE102008056391B4, Ex.1024.

itself can be comprised of  $W_2(hpp)_4$  and BPhen (as exemplar metal compounds and matrix materials). Ex.1005, 58:21–60:14, 61:19–22. Moreover, as discussed above, the POSA would have expected  $W_2(hpp)_4$ :BPhen films to have improved conductivity compared to a pristine BPhen film.

## VI. MA

### A. Ma was published on August 29, 2008

38. I understand that Pictiva disputes that the document of Ex.1007 (the Ma reference) was the version posted online in August 2008. In my opinion, there are none of the hallmarks on Ex.1007 reflecting any substantive modification to the text of Ex.1007 that would suggest that Ex.1007 is any different from the version posted online in August 2008.

39. I am familiar with the publication process of Elsevier journals because many of my research articles have been published in Elsevier journals over the course of my career, including in the 2008–2009 time period. For example, I am the first and corresponding author of an article titled “Organic solar cells with sensitized phosphorescent absorbing layers,” published in a journal called *Organic Electronics* published by Elsevier in 2009. Ex.1027. The cover of this article shows “Article history” under “Article Info,” as reproduced below.



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### Letter

## Organic solar cells with sensitized phosphorescent absorbing layers

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### ABSTRACT

We demonstrate an increase of the exciton diffusion length ( $L_D$ ) of a fluorescent donor layer via the process of sensitized phosphorescence, whereby initially generated singlet excitons are transferred to triplet excitons via a properly chosen dopant molecule. Using a poly(*p*-phenylene vinylene) host doped with the phosphorescent molecule platinum octaethylporphyrin,  $L_D$  increases from  $4 \pm 1$  to  $9 \pm 1$  nm for an optimal doping of 5 wt%. As a result of the increased  $L_D$ , the photocurrent from the doped layer increases by 40%. By doping with the fluorescent, non-metalated analogue octaethylporphyrin, a reduction in photocurrent is shown, providing further evidence of the sensitized phosphorescence mechanism.

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Ex.1027, 1015.

40. Based on my experience, and using my 2009 article as an example, what follows is a rough recitation of the publication process for an article published in an Elsevier journal, such as *Synthetic Metals*. Briefly, once a manuscript is received by an Elsevier journal (see, e.g., *id.*, “Received 2 April 2009”), the journal decides whether it will send the manuscript to reviewers. If the manuscript is sent to reviewers, any reviewers’ comments and questions are sent to the authors, which typically require

revision of the original version of the article to address issues and questions raised by the reviewers. Then the authors revise the manuscript accordingly and send a revised manuscript to the journal (*see, e.g., id.*, “Received in revised form 20 April 2009”). If that version is accepted, the authors receive a letter notifying that the article was accepted (*see, e.g., id.*, “Accepted 21 April 2009”). After the article is accepted, the final version of the article is published online before hard-copy publication (*see, e.g., id.*, “Available online 3 May 2009”). Note that my article, Ex.1027, was published in Volume 10, Issue 5 of Organic Electronics in 2009, the hard-copy journal of which was published in August 2009, 4 months after the online publication in May 2009 (<https://www.sciencedirect.com/journal/organic-electronics/vol/10/issue/5> (last visited July 30, 2025))).

41. I see the same format of “Article history” in Ex.1007, which shows “Received 11 March,” “Accepted 14 May 2008,” and “Available online 29 August 2008” without any “Received in revised form” date or any other indication that there was any revision to the substance to Ma. Thus, there is no evidence to suggest that this article was revised in any way after it was first published online on August 29, 2008. Ma was published as hard-copy in December 2008, 4 months after the online publication, a similar period of time between online and print publication as in my article, Ex.1027, published in 2009.

**B. Joint vaporization of Liq and BPhen is disclosed in Ma**

42. Ma teaches that its electron-conducting layer (“electron transport layer” or “ETL”) is *obtained by joint vaporization* of its metal complex (Liq) with its organic compound (BPhen). Ex.1003, ¶141. I understand that Pictiva argues that “joint vaporization means the two materials vaporize together.” POR, 54. It is my opinion that this is precisely what Ma discloses.

43. Ma describes that “a 50 nm-thick BPhen:Liq ETL was *co-deposited* on the EML.” Ex.1007, 811 (emphasis added). In my opinion, that Ma described the thickness of the two-component ETL with one value suggests that the two components were vaporized together, as opposed to creating a “macroscopically homogenous form” without “the two plumes overlapped with each other,” as Pictiva argues. POR, 54–55.

44. In my opinion, Dr. Schanze’s arguments in relation to a hypothetical deposition chamber for a “macroscopically homogenous” but potentially microscopically heterogeneous film is completely irrelevant to Ma. Ex.2041, ¶¶155–157. The purpose of Ma was to assess the current density using different percentages of Liq doping. The authors would have no reason to prepare a film that is only “macroscopically homogenous” but microscopically heterogeneous across the different locations of a test substrate because it would introduce excessive variability and render the data noncomparable and unreliable.

45. Ma further discloses that that “[t]he deposition rate was *maintained* at

0.5±0.2 Å/s for the organic materials” and that it used “two independent quartz sensors” to monitor the “deposition *rates* for the co-deposition layer.” Ex.1007, 811 (emphasis added). As I testified, this means, the rates of neither vapor streams dropped to zero, and thus were not, for example, sequentially deposited (i.e., not vaporized together). Ex.2042, 81:11–82:7 (“A. During the time of the deposit of the film formation that the rates . . . not going to zero.”). Thus, in my opinion, a POSA would understand Ma’s disclosure of a “BPhen:LiQ ETL [that] was *co-deposited*” to teach (or at a minimum strongly suggest) that the ETL was *obtained by joint vaporization* of Liq and BPhen.

46. Further, it is my opinion that to the extent there is any question whether Ma’s disclosures are sufficient to demonstrate joint vaporization, the POSA would have been motivated to do so with a reasonable expectation of success. “Joint vaporization” is the most natural and straightforward interpretation of “co-deposition” for forming a layer, particularly when there is nothing to indicate that materials were vaporized sequentially or without plume mixing. Because sequential vaporization means two materials are alternately deposited, researchers would specify details of the deposition setup.

47. For example, Yang 2007 [Ex.1028] deposited C<sub>60</sub> and CuPc by sequential deposition. As Figure 1 and its figure description show, the thickness of each alternating layer is specified as “[C<sub>60</sub>(3 nm)/CuPc(3 nm)]<sub>n</sub>,” meaning 3 nm of C<sub>60</sub> and

3 nm CuPc were alternately deposited  $n$  times. Ex.1028, 4167, Figure 1. For example, Yang 2007 used descriptions like, “[CuPc(6.1 nm)/C<sub>60</sub>(6.1 nm)]<sub>10</sub>” and “[CuPc(3.1 nm)/C<sub>60</sub>(3.1 nm)]<sub>17</sub>” to clearly indicate how thick each layer was and how many alternating layers were deposited. *Id.* Figure 1(b) shows how long each material was vaporized and also shows that there was a period of time when only the diluent flowed (shown as the black line) before switching to the other material to prevent any overlap between two materials. Ex.1028, 4167, Figure 1(b). In contrast, Ma discloses that “[t]he deposition rate was *maintained* at  $0.5 \pm 0.2$  Å/s for the organic materials,” which indicates that Ma did not try to separate the Liq and BPhen streams. Ex.1007, 811.

48. By way of another example, Hong 2007 [Ex.1029] discloses sequential deposition of ZnPc and C<sub>60</sub>. Ex.1029, Abstract. Hong 2007 also specifies the thickness of each alternating layer: for example, “[C<sub>60</sub> (1,2,3)/ZnPc(3,2,1)]” for “successively depositing 1 nm C<sub>60</sub>, 3 nm ZnPc, 2nm C<sub>60</sub>, 2 nm ZnPc, 3 nm C<sub>60</sub>, and 1 nm ZnPc,” and “[C<sub>60</sub> (3)/ZnPc (3)] x 2” for 3 nm of C<sub>60</sub> and 3 nm of ZnPc, repeated twice. Ex.1029, 203505-2.

Structure

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- A: ZnPc(20 nm)/C<sub>60</sub>(40 nm)  
B: ZnPc(14 nm)/ZnPc:C<sub>60</sub>(1:1, 12 nm)/C<sub>60</sub>(34 nm)  
C: ZnPc(14 nm)/[C<sub>60</sub>(1, 2, 3)/ZnPc(3, 2, 1)]/C<sub>60</sub>(34 nm)  
D: ZnPc(14 nm)/{[C<sub>60</sub>(3)/ZnPc(3)] × 2}/C<sub>60</sub>(34 nm)  
E: ZnPc(14 nm)/{[C<sub>60</sub>(2)/ZnPc(2)] × 3}/C<sub>60</sub>(34 nm)

Ex.1029, 203505-2, Table 1 (excerpted). Again, in contrast, in Ma, no such statement

of repeating layers is provided, leading to the conclusion that repeated layering did not occur.

49. It is notable that Hong 2007 differentiates a sequentially deposited layer as “a multilayer system” from a “a mixed layer” which is how it annotates “ZnPc:C<sub>60</sub> (1:1, 12 nm).” Ex.1029, 203505-2, Table 1, Structure B. A POSA would understand this annotation as a 12 nm layer formed by joint vaporization of ZnPc and C<sub>60</sub> in a 1-to-1 ratio. This is very similar to how Ma describes the Liq-doped BPhen layer: “BPhen:y wt.% Liq (50 nm).” Ex.1007, 811. There is no other description in Ma that specifies the thickness of each Liq or BPhen layer within the total thickness of 50 nm, or how many times any alternating deposition was repeated, which, again, suggests that it was not deposited in sequential layers.

50. Hong 2007 reports that even though “[t]he total thickness of interlayers or blend layer [were] kept at 12 nm to allow a comparison[,]” each structure showed a different performance. Ex.1029, 203505-2, Table 1. For example, the asymmetric multilayer configuration (i.e., [C<sub>60</sub> (1,2,3)/ZnPc(3,2,1)] offers the best performance among all structures. Ex.1029, 203505-2. Although this work was done for a photovoltaic cell, a POSA would have expected similar variations in performance in an OLED depending on how each alternating layer within an ECL is deposited. Indeed, Dr. Schanze also agreed that “if one did such a . . . sequential deposition, one would expect that it would affect the structure of the material and the device, if it was used in

a device.” Ex.1021, 38:21–39:24. Because of this impact, researchers are expected to note detailed description of alternating layers in the case of sequential deposition.

51. Ma’s description includes no such note detailing alternating layers in the case of sequential deposition, thereby confirming that it was *not* sequential. Ma investigated how the current density changes as the percentage of Liq changes, from 0%, to 17 wt%, 33 wt%, or 50 wt% Liq in BPhen. Ex.1007, 812, Fig. 5. If sequential deposition had been used, the authors would have provided exactly how each Liq and BPhen layer was alternately deposited to dope different percentages of Liq in BPhen (e.g., how many internal layers are deposited, how thick each layer is, whether the thickness of each Liq/BPhen layer varies, etc.). Because Ma does not provide any such intricate deposition setup, a POSA would recognize that Ma vaporized Liq and BPhen together. It is my opinion that joint vaporization would best achieve Ma’s objective of assessing the current density using different Liq doping percentages.

52. I understand that Dr. Schanze argues that Ma does not disclose joint evaporation because “Ma provides no information on its evaporator configuration, the position of the sources, or the degree of intersection, if any, of the plumes from the Liq-source and the BPhen-source.” Ex.2041, ¶154. Dr. Schanze further argues that “co-evaporation” or “co-deposition” does not necessarily mean joint vaporization. Ex.2041, ¶157, n.11. These arguments contradict his own description of joint vaporization. In his own report, Dr. Schanze used the terms “co-deposition” and “co-

evaporation” to indicate his use of joint vaporization in an experiment for the German proceeding. Ex.1026 [Schanze German report], ¶¶48–50; Ex.1021, 37:12–38:20. This report describes the co-deposition method as follows:

Devices:

Evaporations were performed in a vacuum evaporator (MB-EVAP) from MBRAUN. The evaporator was enclosed in a glovebox under an inert atmosphere and was operated at a pressure of  $10^{-9}$  mbar. The film deposition was controlled using an MBRAUN PLC (SIEMENS) unit and an Inficon SQC 310 unit. A sonicator (BRANSON 2800) was used to clean all substrates.

...

c) BTB-LiQ co-deposition

4 cleaned and dried glass substrates were loaded on a substrate holder and mounted in the vacuum evaporator for film deposition. In the evaporating chamber, 50 mg of LiQ and 50 mg of BTB were placed in two separate crucibles with separate heating sources and the chamber was closed and evacuated to a pressure of  $10^{-9}$  mbar. For the BTB-LiQ co-deposited films, the heater power was adjusted in the range 70 – 95% to afford the desired evaporation rate of the two materials. For the BTB-LiQ film with a ratio of 70:30, a rate of 1.4 Å/s for BTB and a rate of 0.6 Å/s for LiQ was used to co-evaporate BTB at  $\sim 290$  °C and LiQ at  $\sim 300$  °C and achieve a film thickness of  $\sim 200$  nm. For the BTB-LiQ film with a ratio of 50:50, a rate of 1 Å/s for BTB and a rate of 1 Å/s for LiQ was used to co-evaporate BTB at  $\sim 280$  °C and LiQ at  $\sim 310$  °C and achieve a film thickness of  $\sim 200$  nm.

Ex.1026, ¶50. I do not see any description on “evaporator configuration, the position of the sources, or the degree of intersection, if any, of the plumes” from the Liq-source and the BTB-source that Dr. Schanze asserts should be included if joint vaporization is used. Indeed, Dr. Schanze admitted that “co-deposition” (or “co-evaporation”) in his report would be understood to refer to joint vaporization, acknowledging that one “would not be able to determine” that he used joint evaporation from the description and that “*if one did such a, ... sequential deposition, ... you would expect it to be stated.*” Ex.1021, 38:21–39:24 (emphasis added); *see also id.*, 36:10–21 (Dr. Schanze referring to his joint vaporization experiments (*see id.*, 38:12-15) as “co-evaporation,” e.g., “The experiments involved the co-evaporation of LiQ and the structure on the left, Figure 2, which you are referring to as ‘BTB’”; “the aim of the experiment was to determine whether a coordination complex forms when LiQ and 742 and co-evaporated. Excuse me. When LiQ and compound BTB are co-evaporated.”).

**C. Description of the manufacturing process in Ki as it relates to Ma**

53. I have been asked to review Dr. Schanze’s report for the German Proceeding (Ex.1026 [Schanze German declaration]) relating to a reference called Ki (Ex.2050). I understand that the patent-at-issue in the German proceeding is the German patent equivalent to the ’223 patent. In this report, Dr. Schanze cited a publication by Ki et al. (Ex.2050, “Ki”) and a publication by Kang et al. (Ex.1030, “Kang”) that cites Ki et al.

54. It is my opinion that Ki used the same materials and the same processing conditions as Ma. In Ki, “OLED devices were fabricated using the mixtures of BPhen and Liq (up to 50%) as a mixed electron transporting layer (mETL) to explore the improvement of device performance upon Liq mixing.” Ex.2050 [Ki], 1677. The Supporting Information of the Ki publication further discloses:

Each film of  $\approx 200$  nm was deposited by a thermal co-evaporation at a base pressure of  $\approx 10^{-7}$  torr. The mixing percentage was controlled from the deposition rates of two source materials, which were individually monitored by two quartz crystal microbalances. The sum of deposition rates of the two materials was kept at  $\approx 2 \text{ \AA s}^{-1}$ .

Ex.1031 [Ki pub, supporting information], 1. In comparison, Ma discloses:

a 50 nm-thick BPhen:Liq ETL was co-deposited on the EML. In the co-deposition process, organic sources were independently controlled. The deposition rates for the co-deposition layer were monitored using two independent quartz sensors. . . . All the layers (organic layers, and Al electrode) of the devices were fabricated by conventional thermal evaporation in a high vacuum chamber with a base pressure of  $1 \times 10^{-5}$  Torr. The deposition rate was maintained at  $0.5 \pm 0.2 \text{ \AA/s}$  for the organic materials, . . .

Ex.1007, 811. In other words, both Ma and Ki provide almost identical descriptions of the vaporization processes, both of which are understood as joint evaporation of Liq and BPhen.

**D. Use of inert gas to manufacture a film of Liq and BPhen in the gas phase**

55. As I discussed in my Opening Declaration, to the extent Ma does not expressly teach coordination of Liq and BPhen in the gas phase, a POSA would have been motivated to modify the parameters of deposition to allow Liq and BPhen to coordinate in the gas phase to obtain a more uniform doping. Ex.1003, ¶165. A POSA would know that a film with uniform doping would be more desirable than nonuniform doping for quality control and reliability of the manufacturing. A POSA would also understand that it would create a more uniform film if molecules coordinate in the gas phase before they impinge on the surface because the mobility of the molecules would be significantly higher in the gas phase than after being fixed on the surface. Ex.2042, 88:8–89:9.

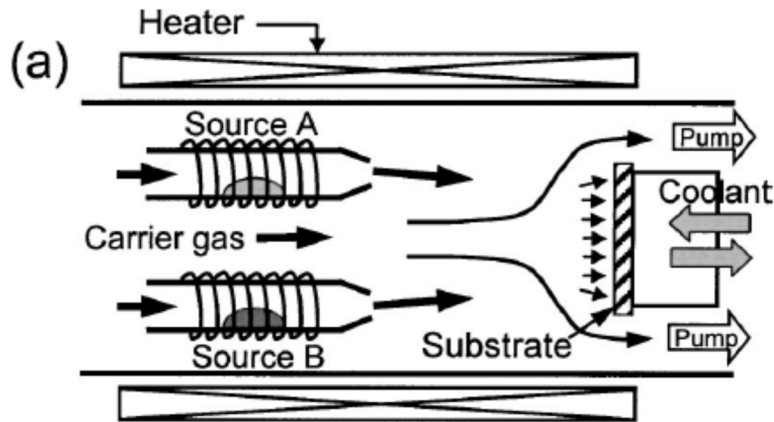
56. I understand that Pictiva argues that “to the extent coordination of BPhen to Liq is possible and coordination increases the uniformity of the film and hence performance, that benefit is equally achievable if the coordination occurs when BPhen and Liq reacts on the deposition surface.” POR, 68. I disagree. Pictiva appears to assume that coordination on the surface would occur as easily as coordination in the gas phase. However, that is not the case. Coordination on the surface means that Liq and BPhen molecules would first impinge on the surface before they collide with each other. Once molecules are adsorbed on the surface, their mobility is significantly reduced compared to the gas phase where they can

freely move and collide with each other, which increases the possibility of coordination. Once molecules are adsorbed, a Liq molecule and a BPhen molecule are not likely to coordinate unless they happen to be adsorbed on the surface in a configuration and at a distance with each other to allow coordination.

57. In contrast, as I explained in my Opening Declaration, “if the BPhen and Liq coordinate in the gas phase, the complex which is deposited will be coordinated BPhen and Liq”, and “a deposited layer of coordinated BPhen and Liq will contain a more uniform distribution of BPhen and Liq, than if the BPhen and Liq were individually deposited such that coordination happens on the surface.” Ex.1003, ¶165.

58. To the extent the Board finds that Ma did not use joint vaporization of Liq and BPhen—which I disagree with—the knowledge of a POSA would have motivated him to modify the conditions with a reasonable expectation of success. Petition, 44–45. As I explained in my Opening Declaration, for example, the OVPD method can be used to increase the possibility of productive collisions between Liq and BPhen molecules. Shtein (Ex.1008) explicitly discloses joint vaporization of two materials (i.e., Source A and Source B) in the presence of an inert gas (i.e. carrier gas), as disclosed in Shtein (Ex.1008, 4006, Fig. 1(a) (reproduced below)). Ex.1003, ¶108. In reference to Fig. 1(a), Shtein states that “[t]he individually heated organic vapors are transported by a carrier gas,” which means that Source A and Source B in Fig. 1

are individually heated. Ex.1008, 4006, Fig. 1(a) caption.



Ex.1008, 4006, Fig. 1(a) (showing Source A and Source B as two separate sources).

59. Dr. Schanze argues that Petitioner provided “no explanation why the presence of large amounts of inert gas molecules would not interfere with the chance that a Liq moiety would collide with BPhen to form coordination” and that “given the complexity of thermodynamics and kinetics, a person of ordinary skill in the art would not have reasonably expected that OVPD would have promoted coordination of Liq with BPhen and resulted in a more uniform film of Liq and BPhen.” Ex.2041, ¶¶190–191. I disagree. In my Opening Declaration, I explained that

a POSA would have understood that when BPhen and Liq are jointly evaporated using the OVPD method, they would likely result in coordination in the gas phase rather than on the substrate because increasing the pressure of the reaction chamber would increase collision of the vaporized Liq and BPhen molecules, which in turn would result in coordination of these compounds in the gas phase.”

Ex.1003, ¶166. A POSA would know that the existence of the inert gas would significantly reduce the mean free path of the molecules, which in turn would increase the possibility of productive collisions.

## **VII. AVAILABILITY FOR CROSS-EXAMINATION**

60. In signing this declaration, I recognize that the declaration will be filed as evidence in a contested case before the Patent Trial and Appeal Board of the United States Patent and Trademark Office. I also recognize that I may be subject to cross examination in the case and that cross examination will take place within the United States. If cross examination is required of me, I will appear for cross examination within the United States during a time as mutually agreed between the parties.

## **VIII. JURAT**

61. I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

I, Dr. Barry P. Rand, declare under penalty of perjury under the laws of the United States of America that the foregoing is true and correct.

A handwritten signature in black ink, appearing to read "B. P. Rand", is written above a horizontal line.

Executed on August 10, 2025