

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

SAMSUNG DISPLAY CO., LTD.,
Petitioner,

v.

PICTIVA DISPLAYS INTERNATIONAL LTD.,
Patent Owner.

Case IPR2024-01095
U.S. Patent No. 11,828,425

**REPLY DECLARATION OF RUSSELL J. HOLMES, PH.D.
IN SUPPORT OF PETITION FOR *INTER PARTES* REVIEW OF
U.S. PATENT NO. 11,828,425**

I. Introduction

I, Russell J. Holmes, Ph.D., declare and state, to the best of my knowledge, understanding, and belief, as follows:

1. I have been retained by counsel for Petitioner Samsung Display Co., Ltd. as an independent expert consultant in this proceeding relating to U.S. Patent No. 11,828,425 (“the ’425 patent”). I am being compensated at my standard hourly consulting rate for my time. My compensation is in no way contingent on the content of my testimony or the outcome of these or any other proceedings relating to the ’425 patent.

2. I previously provided a declaration (Ex. 1003, “Opening Declaration”) providing my expert opinions concerning the validity of certain claims of the ’425 patent in this proceeding.

3. I have been asked to provide this reply declaration in response to certain opinions set forth in the Declaration of Dr. P. Morgan Pattison, PhD. (Ex. 2002) and the Supplemental Declaration of Dr. P. Morgan Pattison, PhD. (Ex. 2018), which I understand were submitted by Patent Owner in this proceeding.

4. My background and qualifications, and my understanding of the relevant legal standards, are detailed in my Opening Declaration. Ex. 1003, §§ II, IV.

5. In forming my opinions, I reviewed the documents in the following table, as well as the documents cited in the Declaration of Dr. P. Morgan Pattison, PhD. (Ex.

2002) and the Supplemental Declaration of Dr. P. Morgan Pattison, PhD. (Ex. 2018),

and other documents cited in this reply declaration.

Exhibit No.	Description
1001	U.S. Patent No. 11,828,425
1002	File History for U.S. Patent No. 11,828,425
1004	Certified Translation of International Patent Application Publication No. WO 2008/040323 A2 (“Diekmann”)
1005	International Patent Application Publication No. WO 2008/040323 A2 (“Diekmann”)
1006	Certified Translation of Japanese Patent Application Publication No. JP 2007-277631 A (“Suzuki”)
1007	Japanese Patent Application Publication No. JP 2007-277631 A (“Suzuki”)
1008	U.S. Patent Application Publication No. 2005/0146267 (“Lee”)
1009	U.S. Patent Application Publication No. 2006/0214892 (“Harada”)
1010	J.W. Ma et al., <i>Organic light-emitting diodes based on new n-doped electron transport layer</i> , Synthetic Metals 158 (2008) (“Ma”)
1011	U.S. Patent Application Publication No. 2003/0159910 (“Caldwell”)
1012	U.S. Patent Application Publication No. 2008/0211395 (“Koshihara”)
1013	M. A. Baldo et al., <i>Highly efficient phosphorescent emission from organic electroluminescent devices</i> , 395 Nature 151 (1998).
1014	M. A. Baldo et al., <i>Very high-efficiency green organic light-emitting devices based on electrophosphorescence</i> 75 Appl. Phys. Lett. 4–6 (1999)
1015	Chihaya Adachi et al., <i>Confinement of charge carriers and molecular excitons within 5-nm-thick emitter layer in organic electroluminescent devices with a double heterostructure</i> , 57 Appl. Phys. Lett. 531–533 (1990)
1016	Martin Pfeiffer et al., <i>Electrophosphorescent p-i-n organic light-emitting devices for very-high-efficiency flat-panel displays</i> , 22 Adv. Mater. 14 (2002)

Exhibit No.	Description
1017	L. S. Hung et al., <i>Enhanced electron injection in organic electroluminescence devices using an Al/LiF electrode</i> , 70 Appl. Phys. Lett., 152–154 (1997)
1018	Zugang Liu et al., <i>Improved electron injection in organic LED with lithium quinolate/aluminium cathode</i> , 128 Synthetic Metals 211–214 (2002)
1019	Gufeng He et al., <i>Very high-efficiency and low voltage phosphorescent organic light-emitting diodes based on a p-i-n junction</i> , 95 J. Appl. Phys., 5773–5777 (2004) (“He”).
1020	L. Moro et al, <i>Process and design of a multilayer thin film encapsulation of passive matrix OLED displays</i> , Proceedings of SPIE Vol. 5214 (2004)
1021	Anna B. Chwang et al., <i>Thin film encapsulated flexible organic electroluminescent displays</i> , 83 Appl. Phys. Lett. 413–415 (2003)
1022	V. Bulovic et al., <i>Transparent light-emitting devices</i> , 29 Nature 380 (1996)
1024	U.S. Patent No. 6,879,319 (“Cok”)
1025	File History for U.S. Patent Application No. 13/139,514
1026	File History for U.S. Patent Application No. 15/651,508
1027	File History for U.S. Patent Application No. 16/790,198
1028	U.S. Patent Application Publication No. 2011/0260201 (“Klein”)
1029	Harald Gross et al., <i>High Power Efficiencies at Record Lifetimes: NOVALED’S PIN-OLEDs</i> , FPD Yokohama 2005
1030	Gufeng He et al., <i>High-efficiency and low-voltage p-i-n electrophosphorescent organic light-emitting diodes with double-emission layers</i> , 85 Appl. Phys. Lett. 3911–3913 (2004)
1031	A. G. Werner et al., <i>Pyronin B as a donor for n-type doping of organic thin films</i> , 82 Appl. Phys. Lett. 4495–4497 (2003)
1032	U.S. Patent No. 5,594,469 (“Freeman”)
1033	M. H. Lu et al., <i>High-efficiency top-emitting organic light-emitting devices</i> , 81 Appl. Phys. Lett. 3921–3923 (2002)
1049	Jun Endo et al., <i>Organic Electroluminescent Devices Having Metal Complexes as Cathode Interface Layer</i> , Jpn. J. Appl. Phys. Vol. 41 (2002)

Exhibit No.	Description
1050	Sung Hoon Choi et al., <i>Improved Efficiency and Lifetime of Organic Light-Emitting Diode with Lithium-Quinolate-Doped Electron Transport Layer</i> , Jpn. J. Appl. Phys. 48 (2009)
1051	Kaushik Roy Choudhury et al., <i>LiF as an n-Dopant in Tris(8-hydroxyquinoline) Aluminum Thin Films</i> , Adv. Mater. 2008, 20
1052	Xinyou Zheng et al., <i>Efficiency improvement of organic light-emitting diodes using 8-hydroxy-quinolinato lithium as an electron injection layer</i> , Thin Solid Films 478 (2005)
1053	Gregor Schwartz et al., <i>Reduced efficiency roll-off in high-efficiency hybrid white organic light-emitting diodes</i> , Appl. Phys. Lett. 92, 053311 (2008)
1054	Gregor Schwartz et al., <i>High efficiency white organic light emitting diodes combining fluorescent and phosphorescent emitter systems</i> , Proc. of SPIE Vol. 6192, 61920Q (2006)
1055	Lumtec, Liq, https://www.lumtec.com.tw/products-view.php?ID=327
1056	Lumtec, LiF, https://www.lumtec.com.tw/products-view.php?ID=371
1057	Commission on Isotopic Abundances and Atomic Weights, Barium, https://www.ciaaw.org/barium.htm
1058	Commission on Isotopic Abundances and Atomic Weights, Cesium, https://www.ciaaw.org/caesium.htm

II. A POSITA's Capabilities and Expectations at the Time of the '425 Patent

6. As discussed in my Opening Declaration, “I have been asked to assume that the earliest possible priority date for the '425 patent is December 11, 2008.” Ex. 1003, ¶ 51. I understand Dr. Pattison has stated that he assumes the same priority date for the '425 patent. Ex. 2002, ¶ 21.

7. As discussed in my Opening Declaration, it is my opinion that “a person of ordinary skill in the art (POSITA) at the time of the alleged inventions would have had a Bachelor's degree in chemistry, physics, or a comparable field of study, and at

least three years of research or industrial experience with organic optoelectronic light-emitting devices. An individual with additional education could have less research or industrial experience and vice versa.” Ex. 1003, ¶ 64. I understand Dr. Pattison has stated that he applies this proposed level of skill in the art. Ex. 2002, ¶ 30.

8. As I have explained in my Opening Declaration, at the time of the ’425 patent, I would have qualified as at least a POSITA. Ex. 1003, ¶ 65. I was actively working in the field at the time, and I had extensive experience in the research of organic electronic materials and devices, and more broadly in the areas of semiconductor physics and materials science.

9. Of particular relevance to the issues in this proceeding, a significant part of my graduate studies included work on *phosphorescent OLEDs*, and I carried out work in the *design, fabrication, characterization, and analysis of phosphorescent blue OLEDs*, realizing record efficiencies, as well as the *first phosphorescent blue OLEDs with exothermic host-guest energy alignment*, and some of the earliest phosphorescent blue OLEDs capable of high efficiency and deep blue color purity. I separately carried out spectroscopic studies of *exciton energy transfer* and *quenching behavior* in organic semiconductor materials and devices to elucidate device operating mechanisms and the interaction of molecular excited states with other excitons and electronic charges, especially under high current excitation. I also contributed to work in the areas of *white phosphorescent OLEDs*, the elucidation of singlet-triplet exciton

spin formation statistics, and the integration of phosphorescent OLEDs with flexible transistors to form display pixels.

10. Since earning my Ph.D. in 2006, I have operated an independent research group in the Department of Chemical Engineering and Materials Science at the University of Minnesota. My group has consistently focused on the study of organic semiconductor materials and optoelectronic devices, with heavy emphasis on organic light-emitting devices (OLEDs), organic solar cells, and the study of excitonic phenomena in organic thin films. Specific to the areas of OLEDs and excitonic phenomena, my research group has demonstrated novel architectures for *phosphorescent OLEDs*, elucidated mechanisms governing *device efficiency and degradation*, and worked to demonstrate methods to engineer *exciton formation, recombination, and migration*.

11. I have advised a number of graduate and undergraduate students in the Departments of Chemical Engineering and Materials Science, Electrical and Computer Engineering, and Chemistry at the University of Minnesota. This includes 20 completed Ph.D. graduate advisees (and an additional 7 current graduate advisees), 5 completed master's degree advisees, and 30 undergraduate advisees. Of my graduated Ph.D. students, several now work in the display and electronics industries.

12. Thus, I have had extensive personal experience as a researcher in the OLED field around the time of the '425 patent, and I have personally trained students who developed into POSITAs around the time of the '425 patent.

13. Dr. Pattison states that a POSITA “would not have been motivated to and could not reasonably expect to just swap materials or dopants into an entirely different stack with different host and emissive materials with a reasonable expectation of success,” and that “[s]uch efforts would require significant experimentation and a large team of cross-disciplinary scientists with far more advanced experience to even begin to accomplish.” Ex. 2018, ¶ 10; Ex. 2002, ¶ 101. I disagree.

14. Dr. Pattison’s assertions about the motivations and expectations of a POSITA in swapping materials and dopants into different OLED stacks are untethered from the reality of how work was conducted in the OLED community at the time of the '425 patent. When an interesting new material or device architecture emerges, there is interest from POSITAs to integrate it into their own work. At the time of the '425 patent, the OLED research community was best described as very high impact and fast moving. Dr. Pattison’s proposed approach of sitting on the sidelines until somebody else figured out the exact physical mechanism by which a new material or architecture functioned was not how POSITAs operated at the time. To the contrary, POSITAs would routinely apply and/or modify materials and architectures whose mechanisms were not fully understood but demonstrated promising device

performance. It was by first building OLEDs and running experiments on those devices that POSITAs would discover the underlying mechanisms governing the behavior of OLED materials and architectures. This approach is reflected in much of the literature cited in this proceeding, which are discussed later in this declaration. To illustrate with some examples:

- Ex. 1010 (Ma) at 811, Figs. 2-3 (studying effect of Liq-doped electron transport layer (ETL) by forming test OLEDs at various doping concentrations and comparing against an undoped OLED as a control).
- Ex. 1016 (Pfeiffer, 2002) at 1634-1636, Figs. 1, 3-4 (studying effect of doping charge-transport layers on phosphorescent OLED (PHOLED) performance by forming OLEDs with Li-doped ETLs and an OLED with LiF EIL, and comparing against undoped PHOLED as control).
- Ex. 1018 (Liu, 2002) at 211-212, Figs. 1, 2 and 4 (studying mechanisms of Liq by forming test OLEDs with Liq EIL and comparing against OLEDs with Ca EIL or LiF EIL as controls; also studying effect of different cathodes by forming test OLEDs with Liq EIL and varying metals used as cathode).
- Ex. 1019 (He, 2004) at 5773-5774, Figs. 1, 3-4, Table 1 (studying the effect of an n-doped ETL in *p-i-n* OLEDs by comparing test OLEDs with Bphen doped with Cesium as ETL against *p-i-i* OLEDs with undoped

Bphen and an EIL of LiF as a control; also studying effect of emission layer thickness on quantum efficiency, power efficiency, and operating voltage of *p-i-n* OLEDs by forming *p-i-n* OLEDs with varying emission layers).

To suggest that a POSITA would have needed to fully understand the mechanism of a material in order to be motivated to use that material or reasonably expect success in constructing an OLED with that material is inconsistent with the practices reflected in the OLED literature.

15. Further, many of the most important innovations in OLEDs were coming from academic laboratories staffed with graduate students, and not exclusively from “large teams of scientists.” Notable examples were the discovery of phosphorescent OLEDs at Princeton University and the demonstration of polymer OLEDs by the University of Cambridge. Research in academic groups often led to related or spin-out companies working in OLEDs. For the aforementioned groups, the companies Universal Display Corporation and Cambridge Display Technologies are notable related and/or spin-out companies.

III. Grounds II and VII: Claims 2, 17 Are Rendered Obvious By Diekmann and Ma, and Suzuki and Ma

16. As discussed in my Opening Declaration, Diekmann in view of Ma renders obvious claims 2 and 17 obvious. Ex. 1003, ¶ 139-147; ¶ 224-229.

17. I understand Dr. Pattison disagrees, arguing that a POSITA would not have used Liq as a dopant in either Diekmann's or Suzuki's electron transport layers based on Ma at the time of the '425 patent. Ex. 2018, ¶12.

A. Liq acts an n-dopant in Ma, and Liq acts as an n-dopant when it is doped into the ETLs of Diekmann and Suzuki

18. Dr. Pattison argues that “a skilled artisan would not have abandoned the proven n-doping mechanism of Diekmann and Suzuki—electron donor n-dopants at significantly lower concentrations (e.g., cesium, barium, and LiF)—for a heavy Liq dopant that appears to affect conductivity through a different mechanism (that is, affecting hopping length and electron mobility).” Ex. 2018, ¶ 27. I disagree.

19. Dr. Pattison seems to acknowledge that Liq can act as an n-dopant. He states that the “Liq dopant ... appears to affect conductivity,” Ex. 2018, ¶ 27, and refers to “Liq as an ETL dopant.” Ex. 2002, ¶ 100. He also explains that the literature (Kumar, Ex. 2004) described Liq as being “used as a dopant of Bphen and Alq3 electron transport materials,” and referred to this as “n-type doping of organic semiconductors” Ex. 2002, ¶ 103. Dr. Pattison also acknowledges that “in Ma, Liq helps improve conductivity,” and that “Ma described BPhen as n-doped.” Ex. 2002, ¶ 112.

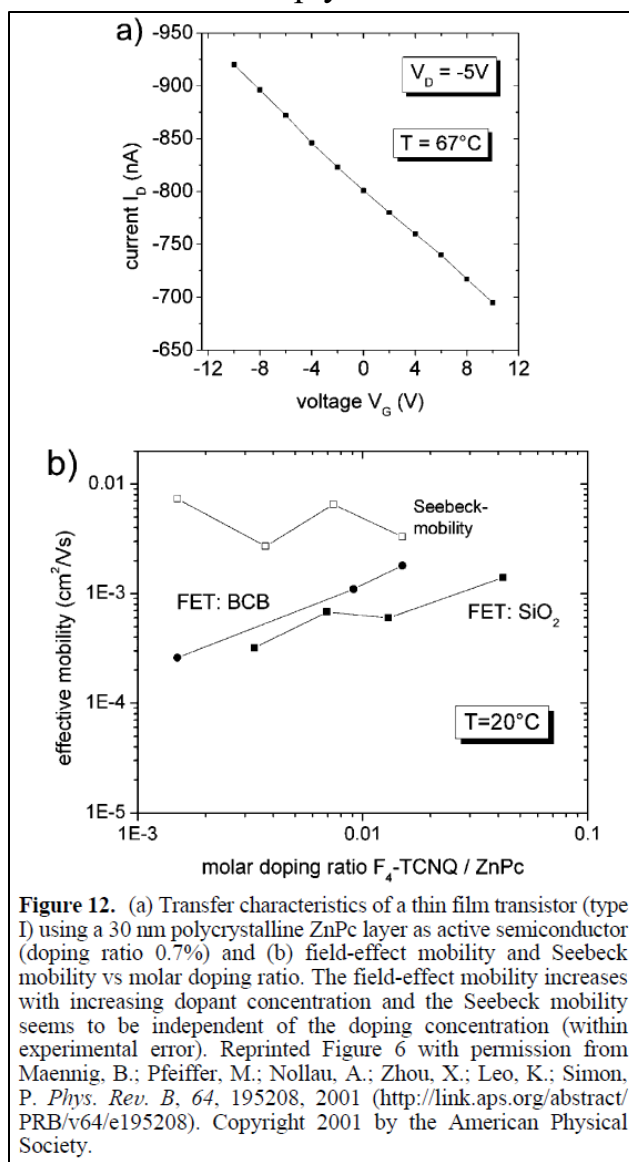
20. Nevertheless, Dr. Pattison argues that a POSITA “would not have chosen Liq in light of Ma because Ma's Liq does not operate in the same way as the electron-donor type n-dopants in Diekmann and Suzuki.” Ex. 2018, ¶ 25. Dr. Pattison thus appears to believe that the mechanisms of improving conductivity by modulating

electron mobility (e.g., through electron hopping) and improving conductivity by increasing electron concentration (e.g., through electron donors) are incompatible.

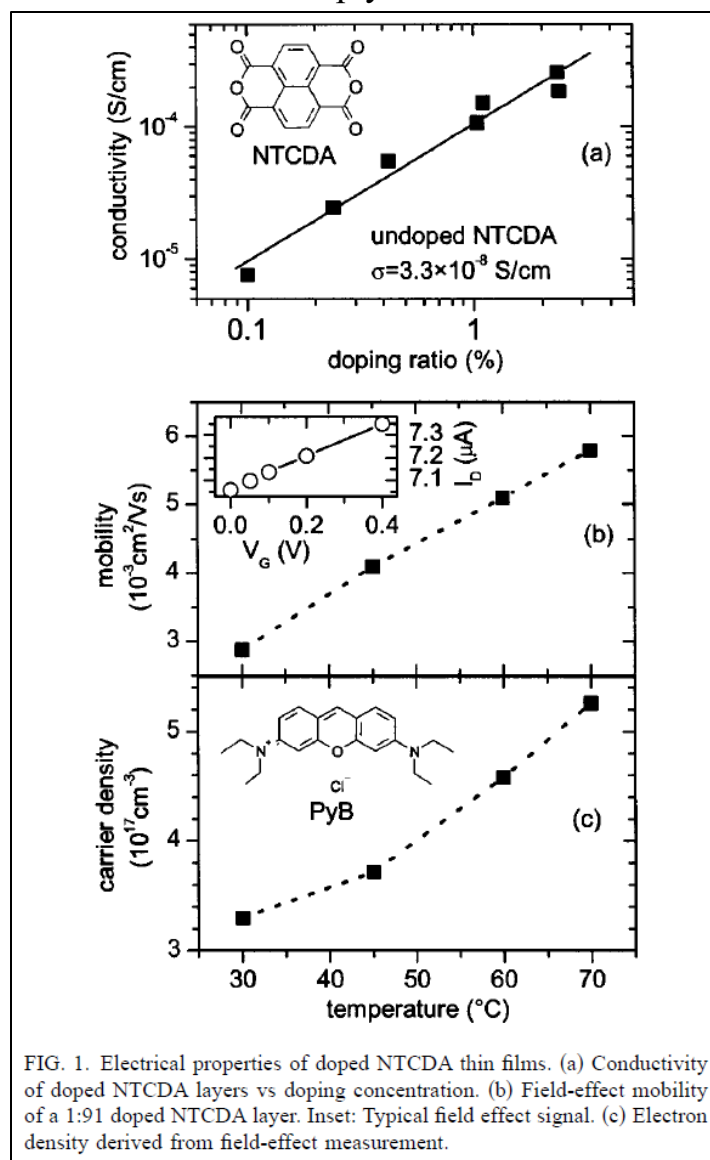
21. I disagree that a POSITA would not have used Liq as the n-dopant in Diekmann's electron transport layer or Suzuki's electron transport layer just because Ma's hypothesized mechanism for how Liq improves conductivity (e.g., electron hopping) is different from the purported mechanism for how some n-dopants in Diekmann and Suzuki improve conductivity (e.g., electron donor). This division in functionality is unnecessary and inconsistent with the prior art.

22. The fundamental purpose of doping charge transport layers in an OLED is to improve the ability of the charge transport layers to bring charge carriers from the OLED electrodes to the emissive layer to generate light. By the time of the '425 patent, it was well-understood that the conductivity of the charge transport layers could be improved by either increasing the number of charge carriers or increasing the mobility of the charge carriers, because conductivity is proportional to the product of carrier mobility and carrier concentration. Dr. Pattison seems to agree on this point. Ex. 2018, ¶ 23 ("conductivity of a layer is the product of carrier concentration and mobility"). But regardless of whether conductivity is improved by changing charge carrier concentration or charge carrier mobility, it is ultimately the conductivity of the charge transport layers that affects key performance characteristics of the OLED, including for example, current density, driving voltage, efficiency, and lifetime.

23. Further, carrier concentration and carrier mobility are not independent parameters. Changes in carrier mobility often occur in tandem with changes in carrier concentration induced by the introduction of charge donor dopants. For example, as noted in Walzer et al., describing prior work by Maennig et al., the p-doping of ZnPc with F4-TCNQ leads to both increased carrier density and increased field-effect mobility. Maennig et al. further notes that “traps in the material become gradually filled [with increased doping]. As a result, the additionally induced charge carriers have a lower probability to be trapped.” Thus the effective mobility is increased. Ex. 2101 (Walzer, 2007) at 1242-1243, Fig. 12.



24. A similar result was reported by Werner et al., for the n-doping of pyronin B into NTCDA. Indeed, the mobility and carrier density increase together with temperature in doped films, both contributing to increased conductivity. Ex. 1031 (Werner, 2003) at 4496, Fig. 1.



25. Thus, a POSITA would not view increases in carrier concentration and increases in carrier mobility due to doping to be distinct phenomena, but rather two related mechanisms by which the conductivity of the transport layer can be improved by doping.

26. Ma also supports this understanding. As Dr. Pattison acknowledges in his declaration, Ma specifically calls out its p-doping mechanism as involving a “charge

transfer” from the HOMO of the matrix material (m-MTDATA) to the LUMO of the p-dopant (F4-TCNQ), and calls out its n-doping mechanism as possibly originating from “short hopping length for electron transport.” Ex. 2018, ¶ 20-21; Ex. 1010 (Ma) at 812-813. The fact that Ma referred to its Liq-doped electron transport layer as an “n-doped electron transport layer,” despite describing the hypothesized mechanism for Liq-doping only in terms of improving electron mobility and not in terms of charge transfer or increased electron concentration, shows that a POSITA’s understanding of what it means to be an “n-dopant” or to be “n-doped” was more expansive than Dr. Pattison suggests, and was not limited to the charge transfer mechanism. Additionally, it is notable that Ma is only *hypothesizing* that “short hopping length” is the mechanism for increased conductivity:

These J–V characteristics of electron-only devices suggest that a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer. The advantage of using BPhen:Liq as ETL can be explained by electron hopping exchange alongwith their LUMO. In a single host device, electrons hop along the LUMO in BPhen. Since the LUMO–LUMO difference between BPhen (3.0 eV) [29] and Liq (3.1 eV) [28] is neglectable, subject to their similar LUMO. Transport manifolds alongwith their LUMO are expected to exhibit a certain extent of overlap after a mixing ratio goes beyond 17 wt.%. Therefore, *it’s likely* that a large energetic disorder between Bphen and Liq contributes to electron hopping, implying that electron hopping among BPhen and Liq sites is favorable [29]. Therefore, the high electron

conductivity of BPhen:Liq *may be originated* from short hopping length for electron transport, compared to the pure BPhen HTL.

Ex. 1010 (Ma) at 813. Ma does not exclude the possibility of contributions from other mechanisms. Indeed, Ma also hypothesizes that the reasons for conductivity decreasing slightly after a certain doping ratio of Liq could possibly be the same as the reasons for conductivity decreasing slightly after a certain doping ratio of “charge transfer” p-dopant F4-TCNQ, suggesting at least some similarities in mechanisms between “donor” type dopants and “hopping” type dopants:

However, the current conduction is reduced slightly as the doping ratio further increases to 50 wt.%. This result may be attributed to the effect of carrier quench and generation of defects. The *detailed reasons are possibly the same as that of the heavy doping concentration of F4-TCNQ* into m-MTDATA in the hole-only devices.

Ex. 1010 (Ma) at 813.

27. Dr. Pattison also argues that Liq is not being used as a molecular n-dopant. Ex. 2018, ¶ 22-23. I disagree. The art recognized that Liq (which is a molecule) could be used as an n-dopant for the electron transport material, and Liq was further known to be effective in facilitating both electron transport and electron injection from as early as 2000 and though the time of the '425 patent. For example:

- Ex. 1010 (Ma) at 813: Liq-doped ETL resulted in “high electron mobility of n-doping ETL”

- Ex. 1049 (Endo, 2002) at L800: Doping ETL with Liq “was also effective in facilitating the electron injection from the cathode to the organic layer”
- Ex. 1050 (Choi, 2009) at 062101-1-062101-2: Liq-doped ETL improves electron injection
- Ex. 2027 (Schmitz, 2000) at 3019: Liq has “good electron injection and electron transport properties”
- Ex. 1018 (Liu, 2002) at 212-214: “Liq is an electron transporting material”; Liq “improves electron injection”

The literature does not support Dr. Pattison’s view that Liq is not a molecular n-dopant.

28. Dr. Pattison further errs by characterizing Diekmann’s n-dopants and Suzuki’s n-dopants as solely “electron-donor” type and characterizing Liq as solely “electron mobility” type without reference to the context in which they are used. Ex. 2018, ¶ 19, 21, 24-25. A POSITA would have recognized that the same n-dopant can act in different contexts to increase electron concentration, increase electron mobility, or both, as discussed above. In fact, Liq itself has been used both as a dopant (e.g., in the electron transport layer) and as a neat interface layer (e.g., as the electron transport layer) in the prior art, and the mechanisms by which Liq improves OLED performance in each case can vary accordingly. For example:

- Ex. 1010 (Ma) at 813 (Liq as ETL dopant showing improved electron transport)
- Ex. 1049 (Endo, 2002) at L800 (Liq as ETL dopant showing improved electron injection)
- Ex. 1018 (Liu, 2002) at 211, 213 (Liq as neat EIL showing improved electron injection)
- Ex. 2027 (Schmitz, 2000) at 3012, 3018-3019 (Liq as neat EIL showing improved electron injection)

29. Dr. Pattison's argument that Liq would not act as an "electron-donor" type n-dopant focuses solely on Liq's function when it is used to n-dope the ETL in Ma's stack, rather than considering Liq's function when incorporated into Diekmann's stack and Suzuki's stack. Ex. 2018, ¶ 19-21. Ma hypothesizes that the principal mechanism by which Liq n-dopes (i.e. increases conductivity of) the ETL appears to be through increased electron mobility. Ex. 1010 (Ma) at 812-813. Since in Ma, a layer of LiF already serves as the electron injection layer, the increase in current seen from Liq-doping the ETL can be attributed to increased conductivity in the ETL, rather than electron injection from the cathode. Ex. 1010 (Ma) at 811. But this does not suggest that Liq-doping would not result in electron injection generally. Indeed, when incorporated as the n-dopant in the electron transport layer of Diekmann's OLED stack or Suzuki's OLED stack, the Liq-doped electron transport layer would be directly

adjacent to the cathode, such that the Liq would be expected to improve electron injection. *See* Diekmann, Fig. 2 (doped layer 402); Suzuki, Fig. 9 (organic layer 7). Notably, an n-doped electron transport layer does not require a separate electron injection layer due to the dual impact of electron transport layer doping on conductivity and injection. The prior art literature describes the mechanism as a form of activated doping, where the Liq dopants are activated by deposition of the cathode:

- Ex. 1049 (Endo, 2002) at abstract, L801-L802: Endo explains that “doping the organic electron-transporting layer with alkaline metal complexes was also effective in facilitating the electron injection from the cathode to the organic layer.” (abstract). One of the metal complexes studied was Liq. Endo et al. explains that “alkaline metal ions in the complexes are reduced to free metal by thermally activated Al. Then, the free alkaline metal reacts with electron-transporting materials, forming the radical anions, which facilitate electron injection from the Al cathode.” (abstract). Endo et al. calls this “an *in situ* chemical doping process.” (L801).

As further explained by Liu et al., Liq dissociates to act like a charge transfer dopant when exposed to the cathode metal. This effect would also be present when Liq is used to n-dope an ETL adjacent the cathode:

- Ex. 1018 (Liu, 2002) at abstract, 212-214: Liu explains that Liq, “covered with aluminum, was used as an electron injection layer.” (abstract). “An Liq electron injecting layer has also been tried in combination with Ca, Mg and Ag cathodes. Our experiments support the assumption that free lithium is released from lithium quinolate, as in the case of lithium fluoride, when Liq is over coated with active metals such as Al.” (abstract). Liu reports “improved electron injection.” (211). Liu explains that “lithium quinolate reacts partially with Ag, to give free lithium that improves electron injection.” (213). Liu further notes that “experimental results for Ca, Mg and Ag cathodes support the assumption that free lithium released from lithium quinolate is responsible for the improved device performance. (214).

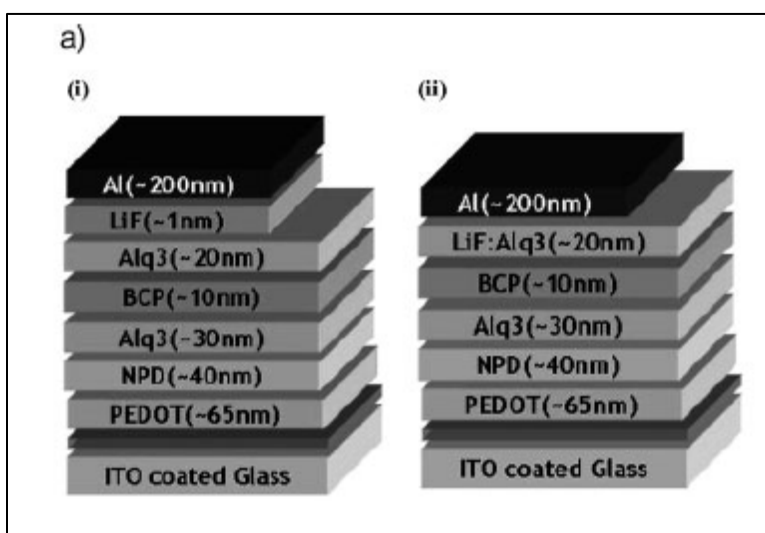
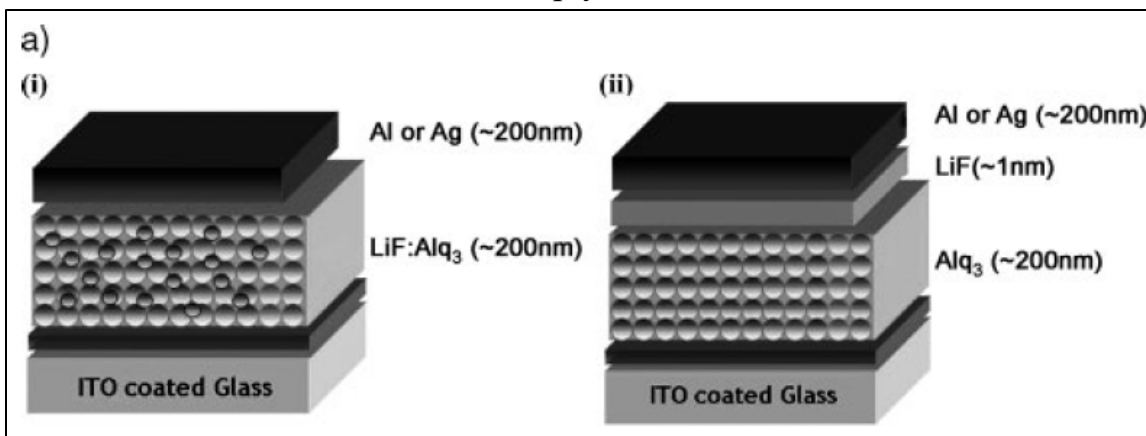
Thus, Liq would act as an “electron-donor type n-dopant,” under even Dr. Pattison’s narrow view of what n-doping requires (Ex. 2018, ¶ 17, 23-27), when Liq is used as the ETL n-dopant in either the Diekmann OLED stack or the Suzuki OLED stack.

30. Further, a POSITA would understand that the purported “electron-donor” n-dopants and Liq are used in analogous contexts for the same reasons, supporting their interchangeability. As discussed above, Liq has been used as both a dopant in the electron transport layer and as a neat interface layer, as exemplified by the various uses of Liq in the literature. For example:

- Ex. 1010 (Ma) at 813 (Liq as ETL dopant)
- Ex. 1049 (Endo, 2002) at L800 (Liq as ETL dopant)
- Ex. 1018 (Liu, 2002) at 211, 213 (Liq as neat EIL)
- Ex. 2027 (Schmitz, 2000) at 3012, 3018-3019 (Liq as neat EIL)

Likewise, Diekmann shows that barium and lithium fluoride (LiF) can be used as both n-dopants for the electron transport layer and neat electron injection layers. Ex. 1004 (Diekmann) at 5 (listing “LiF,” “Ba” as exemplary materials for “the electron injecting layer”), 10 (“Preferred n-dopants are cesium, barium and lithium fluoride” for increasing the conductivity of the doped organic layer adjacent to the cathode.). That Liq and these “electron-donor” n-dopants are used for the same purposes in the same types of layers in the prior art literature further supports that a POSITA would not have considered them to be disparate categories of n-dopants, but instead analogues that work through similar mechanisms.

31. For example, Choudhury et al. shows comparative structures with both LiF as EIL and LiF as ETL dopant. Ex. 1051 (Choudhury, 2008) at 1456, 1460, Fig. 1 (single-carrier devices), Fig. 4 (OLED).



Choudhury also confirms that the mechanism for LiF doping is *in situ* doping activated by deposition of the cathode—the same as the mechanism for Liq doping shown in Endo, discussed above. Ex. 1051 (Choudhury) at 1459 (“Once Al atoms reach the bulk of the Alq₃ layer, LiF molecules might be “activated”[18] and become active dopants in the Alq₃ film.”). Choudhury further notes that LiF doping resulted in “higher carrier mobility in the doped samples compared to the undoped samples,” and concludes that “doping leads to a gradual filling of intrinsic charge traps in the material, resulting in higher low-field carrier mobility compared to undoped samples.” Ex. 1051

(Choudhury) at 1457, 1460. These findings of increased mobility in the electron transport layer due to LiF-doping are further consistent with Ma's findings of increased mobility in the electron transport layer due to Liq-doping, as discussed above. *See also* Ex. 1049 (Endo, 2002) at L800, L802 (LiF as ETL dopant); Ex. 1018 (Liu, 2002) at abstract, 211, 212-213 (LiF as EIL).

32. In particular, Dr. Pattison calls LiF an “electron donor” n-dopant. Ex. 2018, ¶ 15-17, 27. It is well-established that the mechanism by which LiF improves electron injection is analogous to the mechanism by which Liq improves electron injection. Ex. 1018 (Liu, 2002), 213 (lithium released from LiF/Liq dopes organic layer); Ex. 1052 (Zheng, 2005), 254 (electron injection enhancement with Liq “very similar” to LiF). A POSITA would therefore have understood that when Liq is used as an n-dopant in an ETL of an OLED, the Liq would be acting as an “electron donor” n-dopant in the same way that LiF would acting as an “electron donor” n-dopant when the LiF is used as an n-dopant in the ETL of the OLED. Thus, a POSITA would have understood Liq is a suitable n-dopant to use in place of n-dopants like LiF, and further would have reasonably expected success in creating OLEDs with Liq-doped ETL, just as others have succeeded in creating OLEDs with LiF-doped ETL. It is well within the skill of a POSITA to swap out one n-dopant in the ETL for another n-dopant where the doping mechanisms are similar.

33. Even if one were to assume that Liq acts only as an “electron mobility” type n-dopant that operates differently from what Dr. Pattison calls the “electron-donor” type n-dopants when incorporated into the Diekmann OLED and the Suzuki OLED, using Liq dopant in place of electron-donor dopants does not amount to a fundamental alteration of the principle of operation of the OLED. The basic principle of operation of the OLED is that charge transport layers bring charge carriers from the electrodes (electrons from the cathode, and holes from the anode) into the emissive layer to generate light. Regardless of how conductivity in a charge transport layer is improved (through mobility or carrier concentration), the basic principle of operation of the OLED remains the same. Thus, the proposed combinations that simply replace n-dopants with Liq to perform the same function of doping the ETL do not fundamentally alter the principle of operation of Diekmann or Suzuki.

B. A POSITA would have expected that incorporating Liq would have resulted in reduced dopant diffusion and higher efficiency

34. Dr. Pattison argues that there is no direct evidence of reduced diffusion or higher efficiency and lower driving voltages when using Liq as the n-dopant. Ex. 2018, ¶¶ 29, 31, 33. But a POSITA would not have needed to see direct experimental comparisons between Liq and Diekmann’s preferred n-dopants to understand that Liq would have resulted in an improvement. Even absent direct measurement of the relative rates of dopant diffusion or efficiency and driving voltage, the prior art

provided sufficient motivation for a POSITA to use Liq. Diekmann taught that it is “usually the case” that “large atoms or molecules” have “a low diffusion constant within the organic layer stack.” Ex. 1004 (Diekmann) at 10. Dr. Pattison does not dispute this. As I have explained in my Opening Declaration, Liq has a larger molecular weight than Diekmann’s preferred n-dopants. Ex. 1003, ¶ 144. Dr. Pattison concedes that “Liq might have a larger molecular weight.” Ex. 2002, ¶ 82. To avoid any doubt, the molecular weight for Liq, LiF, Barium, and Cesium are as follows:

- Liq: 151.09 g/mole. *See* Ex. 1055 (Lumtec, Liq, <https://www.lumtec.com.tw/products-view.php?ID=327>);
- LiF: 25.94 g/mole. *See* Ex. 1056 (Lumtec, LiF, <https://www.lumtec.com.tw/products-view.php?ID=371>);
- Barium: 137.327(7) g/mole. *See* Ex. 1057 (CIAAW, Barium, <https://www.ciaaw.org/barium.htm>);
- Cesium: 132.90545196(6) g/mole. *See* Ex. 1058 (CIAAW, Cesium, <https://www.ciaaw.org/caesium.htm>).

A POSITA thus would have recognized Liq would be advantageous in reducing dopant diffusion.

35. A POSITA would further have recognized that Liq could have additional benefits over n-dopants like Diekmann’s preferred n-dopants, and would not view existing dopant materials contemplated by Diekmann or Suzuki as the final, optimized

solution. For example, Zheng et al. noted that “[t]o achieve a balanced injection of carriers, it is common to use low-work-function metals such as Li, Ca, Mg, etc., to enhance electron injection into organic materials. However, such metals are not stable in air and need to be protected in the device processing, and sometimes may react with and diffuse into organic material and deteriorate the device.” Ex. 1052 (Zheng, 2005) at 252. Thus, POSITAs would have expected Liq to be more stable than low-work-function metals, such as cesium and barium, when used to n-dope an ETL. Zheng et al. further noted that insulating materials like LiF “need quite high temperature for evaporation and accurate control of thickness.” Ex. 1052 (Zheng, 2005) at 253. Liu et al. also taught that Liq can be made thicker than LiF without significantly compromising EL characteristics such as power efficiency, because Liq has electron transporting properties while LiF is insulating and can blocks electron injection at greater thicknesses. Ex. 1018 (Liu, 2002) at 212-213. Thus, POSITAs would have expected that Liq would be easier to process than LiF when used to n-dope an ETL.

36. Diekmann discloses an n-doped layer and explains that large dopant size is one factor for selecting preferred dopants. Ex. 1004 (Diekmann) at 10. Liq is one such dopant, and the fact that Diekmann also considered other dopants to be good candidates does not make using Liq non-obvious. Diekmann is not limited to its preferred dopants, and does not teach away from other dopants. Ex. 1004 (Diekmann) at 91 (“dopant 410 can be *for example* cesium, barium or lithium fluoride”). A

POSITA thus does not need data showing Liq is superior to the preferred dopants to be motivated to use Liq. The same is true for Suzuki, which does not prescribe any preferred n-dopants. Ex. 1006 (Suzuki) at [0333].

37. Dr. Pattison cites to a number of recent studies from long after the '425 patent, dating from 2011 to 2023. Dr. Pattison argues that these papers show that an Liq-doped ETL would have reduced OLED performance. Ex. 2018, ¶ 34-39. This post-priority date evidence does not reflect what was known in the art at the time of the '425 patent or what the expectations of a POSITA would have been at the time. Nor is Dr. Pattison's characterization of these studies correct.

38. For example, the study from 2011, which is closest to the priority date and thus most likely to be reflective of a POSITA's understanding at the time, shows that the use of an Liq-doped ETL *improved* efficiency and operating voltage, consistent with a POSITA's expectations at the time of the '425 patent based on Ma's teachings:

In this paper, we demonstrate development of red phosphorescent Organic Light-Emitting Diodes (OLEDs) *as the doping concentrations are varied with lithium-quinolate (Liq) doped electron transport layer (ETL)*. The experimental device has both mixed electron transport layer and electron injection layer. The device configuration was ITO/NPB (50 nm)/4,4'-N-N'-dicarbazole-biphenyl (CBP) doped with 8% bis(2-phenylquinoline)iridium(III) (acetylacetonate) [Ir(pq)₂(acac)] (30 nm)/Bphen doped with X% Liq (30 nm)/Liq (2 nm)/Al (100 nm), where

the doping concentration of Liq in ETL was varied as 5%~65%. ***When the Liq was mixed in the ETL, the efficiency and operating voltage of the experimental devices were improved in comparison with the reference device due to greater electron transport efficiency and an improved hole-electron balance.***

Ex. 2003 (Kim, 2011) at 53, 56-60.

39. Similarly, the next earliest study, from 2013, also shows that Liq-doped ETL showed the best efficiencies, in terms of luminous efficiency, power efficiency, and quantum efficiency, consistent with a POSITA's expectations at the time of the '425 patent based on Ma's teachings:

Systematic studies on carrier injection and transport are very important for achieving high efficiency in OLEDs. We demonstrate excellent green phosphorescent organic light-emitting diodes (OLED) with ***lithium quinolate (Liq) doped in 1,3,5-tris(N-phenylbenzimidazole-2-yl) benzene (TPBi) as the electron transport layer (ETL).*** The doping concentration of Liq was varied from 0% to 10%. ***The optimized green phosphorescent OLED with 5% Liq in the ETL showed the best efficiencies, which were maximum luminous efficiency, power efficiency, and quantum efficiency of 65.76 cd/A, 57.39 lm/W, and 20.03%, respectively.*** Moreover, high triplet energy states of TCTA and TPBi as a triplet exciton-blocking layer (TEBL) played a role in efficient exciton confinement.

Ex. 2024 (Kim, 2013) at 7998, 7999-8001.

40. Dr. Pattison also argues that there is no evidence that using an Liq-doped ETL could lead to an OLED with proper charge balance for Diekmann's or Suzuki's emissive layers with multiple emissive layers and a phosphorescent iridium-based emitter. Ex. 2018, ¶ 40-41. But the claims do not require an OLED with the best or most optimized charge balance. Nor is optimum charge balance required for a functioning OLED. In any case, identifying the appropriate doping concentration for good charge balance is a matter of routine optimization well within the skill of a POSITA. There are many examples just in the literature of record in this proceeding showing how researchers could readily create OLEDs at various different doping concentrations, measure their electrical characteristics, and determine which doping concentrations result in the highest current density and lowest driving voltage. For example:

- Ex. 1050 (Choi, 2009) at 062101-2-062101-3 (measuring different Liq doping ratios)
- Ex. 2101 (Walzer, 2007) at Fig. 6 (varying concentration of F4-TCNQ in ZnPc to change conductivity), Fig. 18 (varying concentration of pyronin in NTCDA to vary conductivity), Fig. 27 (showing impact of doping concentration on current flow through an OLED)

Ma itself also demonstrates that the Liq doping concentration for optimum charge balance can be experimentally determined, Ma, 813, as Dr. Pattison acknowledges. Ex.

2018, ¶ 41. These papers further illustrate that operating OLEDs do not require optimum charge balance, because they show that functioning devices were formed at various different doping concentrations.

41. Dr. Pattison also argues that there are no teachings or suggestions about how to select a suitable ETL, or how to use Liq in a white OLED with an emissive layer having a phosphorescent emitter. Ex. 2018, ¶ 42-43. Given an emissive layer, identifying an ETL material that is appropriate for the emitter materials and that meets the required energy levels for injection and transport is within the skill of a POSITA. POSITAs in the OLED field routinely design OLED stacks based on energy level alignment and known material compatibilities. White OLEDs do not require a different analysis. Indeed, the prior art includes many examples of white OLED stacks with appropriate choices of ETL, dopant type, and concentration capable of being determined experimentally. *See, e.g.*, Ex. 1053 (Schwartz, 2008) at 053311-1, Fig. 1; Ex. 1054 (Schwartz, 2006) at 61920Q-1-61920Q-2, Fig. 1.

42. Dr. Pattison argues that a POSITA would have looked to proprietary materials from Novaled as a starting point rather than experimenting with Liq. Ex. 2018, ¶ 44. I disagree. First, such materials would likely not have been widely available, especially to academic researchers. Second, use of Novaled's n-dopant, which is a proprietary material having an unknown structure, over Liq, with reported structure, characteristics, and performance, presents challenges for future innovation.

It would have been particularly undesirable from a research and development perspective, because using a proprietary material makes it much harder to determine why device performance improved (or deteriorated). This makes further innovation using the material difficult. Further, the mere fact that one material set had favorable performance does not dissuade a POSITA from pursuing modifications of other OLEDs.

43. Dr. Pattison also overreads a passage from my deposition transcript out of context to argue that, if dopant diffusion was already a solved issue at the time of the '425 patent, “that would beg the question of why a skilled artisan would have attempted to substitute preferred n-dopants (Cs, Ba and LiF) with Liq which operated using a different mechanism and whose behavior and performance were less understood.” Ex. 2018, ¶ 32. As I clarified immediately after the quoted passage, blocking layers were already commonly being used in PHOLEDs to allow the doped layer to be spaced from the emissive layer, thus “reducing the concern related to dopant diffusion.” Ex. 2019, 28:18-29:14. Later in the same deposition, I again testified that dopant diffusion was a problem that was “mitigated” by the time of the '425 patent, but it was still an issue to be considered. Ex. 2019, 38:8-39:3, and made clear that blocking layers for preventing dopant diffusion were already well known at the time, such that the issue was “already mitigated or at least reduced” by the time of the patent. Ex. 2019, 39:5-40:10.

C. Liq was not a new or unknown material at the time of the '425 patent, and Liq had known benefits that would have motivated a POSITA to use an Liq-doped ETL

44. Dr. Pattison argues that Liq-doped OLEDs were not well-studied at the time of the '425 patent and that there were no known benefits. Ex. 2002, ¶ 74-77, 67, 87. This is incorrect. Dr. Pattison argues that Ma was the only cited reference showing an Liq-doped ETL, but this ignores that Liq-doped ETLs were well-known and that Liq was known to have good electron injection properties. As I explained in my deposition testimony, the Kido group at Yamagata University worked on doping Liq into an ETL. Ex. 2019, 16:17-18:8. A 2002 paper by this group showed Liq-doped ETLs led to better electron injection and reduced driving voltage, Ex. 1049 (Endo, 2002) at L800, L802, consistent with Ma's results, Ex. 1010 (Ma) at 810, 813. Other prior art also shows Liq's effective electron-injection properties, further noting Liq's doping effect. Ex. 2027 (Schmitz, 2000) at 3012, 3018-3019; Ex. 1018 (Liu, 2002) at 211, 213-214.

45. Dr. Pattison's assertion that POSITAs would not have extended Ma's Liq teachings beyond Ma's own OLED stack, Ex. 2002, ¶ 87, is also contrary to how research was conducted in the field. As I explained during my deposition, POSITAs in the OLED field can review the literature, understand how the teachings could be beneficial, and apply those teachings to make their own devices better. Ex. 2019,

211:3-212:4. A POSITA would have read Ma and would naturally have been motivated to apply its teachings about Liq as an ETL dopant to other OLEDs.

46. This is illustrated by the fact that researchers have used the same dopant for multiple different types of HTL or ETL materials. For example, the p-dopant F4-TCNQ has been used in a host of different HTL materials, including ZnPc (Ex. 2101 (Walzer, 2007) at Fig. 10), m-MTDATA (Ex. 1016 (Pfeiffer, 2002) at 1633), MeO-TPD (Ex. 1019 (He, 2004) at Fig. 1), and TDATA (Ex. 2101 (Walzer, 2007) at Fig. 27). As examples on the ETL side, the n-dopant Li has been used in BPhen (Ex. 1016 (Pfeiffer, 2002) at 1633), BCP (Ex. 2101 (Walzer, 2007) at 1245), and Alq₃ (Ex. 1049 (Endo, 2002) at L802).

47. Similarly, different dopants have been used in the same ETL material used in different OLED stacks. Li was the n-dopant for a BPhen ETL used for a PHOLED (Ex. 1016 (Pfeiffer, 2002) at 1633), as well as for a BPhen ETL used for a fluorescent OLED (Ex. 2101 (Walzer, 2007) at Fig. 29). Cs was the n-dopant for a BPhen ETL used for a PHOLED (Ex. 1019 (He, 2004) at Fig. 1). And Liq has been used as the n-dopant for a BPhen ETL (Ex. 1010 (Ma) at 810-811; Ex. 1049 (Endo, 2002) at L802, Fig. 1).

48. In particular, Endo et al. reported OLEDs including either Liq-doped Alq₃ or Liq-doped BPhen. Ex. 1049 (Endo, 2002) at L800-802; Fig. 1. Thus, although Ma only discusses Liq-doped BPhen, a POSITA would have been motivated to apply Liq

to other ETLs, including the Alq₃ material specifically identified as an ETL material in Diekmann (Ex. 1004, 5 (“Alq₃ = Tris(8-hydroxy[quinoline])aluminium”)) and Suzuki (Ex. 1006, [0331] (“tris(8-quinolinol)aluminum (Alq)”)), and would have reasonably expected success in making such devices.

D. The fact that Diekmann and Suzuki are PHOLEDs does not dissuade a POSITA from using Liq

49. Dr. Pattison argues that a POSITA would not have used Liq to dope the ETL in PHOLEDs like Suzuki and Diekmann, which have phosphorescent emitters, because according to Dr. Pattison, “the research into using Liq in OLED devices was nascent and focused primarily on Liq as an injection layer material in fluorescent OLED stacks, not as an ETL dopant in phosphorescent OLED devices,” and “[t]here is no iridium-containing emission layer in Ma’s fluorescent system.” Ex. 2002, ¶ 117-120. I disagree.

50. That Diekmann’s and Suzuki’s OLEDs use phosphorescent emitters does not dissuade a POSITA from using Liq as an ETL n-dopant in view of Ma’s teachings, and a POSITA would have had a reasonable expectation of success.

51. While phosphorescent and fluorescent emitters have some differences in light emission mechanism, there is not a fundamental difference between phosphorescent and fluorescent OLEDs that would prevent a POSITA from applying teachings from one class of OLEDs to the other. To the contrary, by the time of the ’425 patent, researchers routinely looked to teachings of fluorescent OLED literature

to apply to phosphorescent OLEDs and vice versa. And because fluorescent emitter materials were applied in initial demonstrations of thin film OLEDs, POSITAs routinely borrowed from materials and techniques used in fluorescent OLEDs when developing phosphorescent OLEDs.

52. The literature reflects this cross-development in the similar structure of OLED stacks used for both fluorescent and phosphorescent OLEDs, as well as the application of similar materials or layers in both OLED types. For example, the 2007 Walzer paper explains how the “p-i-n architecture” was first developed for fluorescent OLEDs, and then “extended to phosphorescent OLEDs.” Ex. 2101 (Walzer, 2007) at 1254-1255. The fluorescent OLED structure (Fig. 29) is nearly identical to the phosphorescent OLED structure (Fig. 30) except the phosphorescent emitter replaces the fluorescent emitter and a different blocking layer is used. Ex. 2101 (Walzer, 2007) at 1254-1255; Ex. 1016 (Pfeiffer, 2002) at 1633-1634. Notably, the n-doped ETL Li:Bphen is used for both fluorescent and phosphorescent OLEDs. Ex. 2101 (Walzer, 2007) at 1254-1255; Ex. 1016 (Pfeiffer, 2002) at 1633-1634. This demonstrates the modular approach to OLED stack design, even across different emitter types, and shows that it was not a big leap for a POSITA to apply ETL n-dopants studied in the context of fluorescent OLEDs, such as Ma’s Liq, as the n-dopant for PHOLEDs with n-doped ETLs.

53. Further, n-doping the electron transport layer was a well-known technique for both fluorescent and phosphorescent devices by the time of the '425 patent, and some of the same n-dopants have been used in both fluorescent and phosphorescent devices. Ex. 2101 (Walzer, 2007) at 1254-1255. In fact, OLEDs that have both fluorescent and phosphorescent emitters as part of the same OLED and sharing the same organic stack layers had also been developed before the date of the '425 patent. Ex. 1053 (Schwartz, 2008) at 053311-1, Fig. 1; Ex. 1054 (Schwartz, 2006) at 61920Q -1-61920Q-2, Fig. 1.

54. A POSITA would have reasonably expected success in applying Liq, which was found to have favorable performance in the context of Ma's fluorescent OLEDs, to Diekmann's and Suzuki's PHOLEDs with n-doped ETLs. That Diekmann and Suzuki have multiple emission layers does not result in any increase in complexity in making the proposed modification, which is merely using Liq as the n-dopant in a stack that already contemplates an n-doped ETL. One current flows through the OLED stack regardless of the number of emitters, and the charge would be balanced for the emission layer as a unit in the same manner that charge can be balanced for a single-emitter OLED. Selecting the appropriate dopant concentrations and layer thicknesses to optimize charge balance was well within the capability of a POSITA. For example, Ma demonstrates how dopant concentration can be varied to experimentally determine the optimized doping level for charge carrier balance. Ex. 1010 (Ma) at 813. Other

literature also supports that it was well within the POSITA's skill to identify the optimal Liq concentration in any given OLED stack. For example:

- Ex. 1050 (Choi, 2009) at 062101-2-062101-3 (varying Liq doping ratios)
- Ex. 2101 (Walzer, 2007) at Fig. 6 (varying concentration of F4-TCNQ in ZnPc to change conductivity), Fig. 18 (varying concentration of pyronin in NTCDA to vary conductivity), Fig. 27 (showing impact of doping concentration on current flow through an OLED)

55. Dr. Pattison also argues that using Liq as the n-dopant in a PHOLED would have resulted in significant problems because Liq can act as a luminance quencher, and can dissociate to generate free lithium that could diffuse through the OLED and interact with the triplet excitons to cause them to lose energy through non-radiative pathways. Ex. 2018, ¶ 47-53, Ex. 2002, ¶ 38-39, 44, 82, 106-107, 115, 121. I disagree.

56. The potential for luminance quenching is not an issue unique to Liq, and thus does not dissuade a POSITA from using Liq as the n-dopant. Many dopant species would be expected to undergo some amount of diffusion within the stack. For example, any of Diekmann's preferred n-dopants (Cs, Ba, LiF) could also act as luminance quenchers. Further, as I have explained above, the mechanism by which Liq could undergo dissociation would be understood to be similar to that for LiF, one of

Diekmann's own preferred dopants. Ex. 1004 (Diekmann) at 10. A POSITA would thus have expected that Liq would also be an effective n-dopant.

57. Dr. Pattison cites to a 2008 paper by Lee purportedly noting that the binding energy between Li and quinolate is weaker than that of LiF, and that in some forms of Liq, the lithium cation is more easily dissociated and diffused. Ex. 2018, ¶ 49. Even if Liq were more easily dissociated than LiF, a POSITA would not have expected significant quenching to result. The art has long used blocking layers placed between the charge transport layers and the emissive layer to mitigate exciton quenching, and this has been successfully implemented specifically in the context of OLEDs using doped charge transport layers. Ex. 1016 (Pfeiffer, 2002) at 1633-1634, Fig. 1 (blocking layers with Li-doped ETL); Ex. 1019 (He, 2004) at 5773-5774, Fig. 1 (blocking layers with Cs-doped ETL); Ex. 1030 (He, 2004) at 3911-3912, Fig. 1 (blocking layers with Cs-doped ETL). Indeed, because Liq is a larger molecule with a higher molecular weight, a POSITA would understand it would be less mobile than these elemental n-dopants like lithium or cesium. *See also* Ex. 1004 (Diekmann), 10 (“the dopant preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example”). Thus, that blocking layers were sufficient to alleviate quenching concerns for n-dopants like lithium and cesium supports a POSITA's expectation that blocking layers would be sufficient to alleviate quenching concerns for Liq used as an n-dopant.

58. In fact, in Pfeiffer, blocking layers provided “nearly complete isolation of the EML from the conductive dopants,” even where the n-dopant was pure elemental lithium Ex. 1016 (Pfeiffer, 2002) at 1635. A POSITA thus would not have expected that n-doping with Liq, even if dissociated and even if the degree of dissociation were more significant than other dopants like LiF, would result in significant quenching, because consistent with typical PHOLED structures in the prior art, both Diekmann and Suzuki already include blocking layers. Ex. 1004 (Diekmann) at Fig. 2A; Ex. 2019, 185:20-186:5; Ex. 1006 (Suzuki) at [0287], [0296]-[0298].

59. As discussed above, blocking layers could adequately address quenching issues with a variety of n-dopants, and this technique was well-known in the art by the time of the '425 patent. Thus, a POSITA would not have been dissuaded from pursuing Liq n-dopants with Diekmann's or Suzuki's OLED stacks merely because of a possibility of some quenching behavior. This is further evidenced by the prevalence of other doped PHOLEDs, such as those discussed above, and the fact that prior researchers similarly took n-dopants used in fluorescent devices and successfully applied them to PHOLEDs, and even using almost identical architectures for both the fluorescent and phosphorescent devices. *See, e.g.*, Ex. 2101 (Walzer, 2007) at 1254-1255, Fig. 29-30.

E. There was no reason to believe that using Liq n-dopant in Suzuki or Diekmann's ETL would create a charge imbalance in the OLED stack

60. Dr. Pattison argues that Liq doping “could” result in “charge imbalance due to an electron surplus and undesirable traps.” Ex. 2018, ¶ 54-55. But Dr. Pattison has not identified any evidence of charge balance in Suzuki and Diekmann, let alone how that would change when the ETL is Liq-doped. As a matter of routine optimization in building the OLED, a POSITA could, and would, choose appropriate p-/n-doping concentrations and layer thicknesses for charge balance and efficiency.

61. Dr. Pattison again relies on a series of studies that post-date the '425 patent to allege that Liq's properties could result in charge imbalance. Ex. 2018, ¶ 54-56. This post-priority date evidence is indicative of the knowledge or expectations of a POSITA at the time of the '425 patent. And as I have explained above, identifying the appropriate doping concentration for optimum charge balance is a matter of routine optimization within the skill of a POSITA, and Ma shows that a POSITA can determine the Liq doping concentration for good charge balance. Ex. 1010 (Ma) at 813.

62. Dr. Pattison alleges that “significant work continued” after the time of the '425 patent to determine “correct energy band levels to dope materials with Liq for an operable system.” Ex. 2018, ¶ 56. But Dr. Pattison ignores that operable systems were already present in the prior art. *See* Ex. 1010 (Ma) at 810-811; Ex. 1049 (Endo, 2002)

at L802, Fig. 1. The single cited paper does not support Dr. Pattison. That multiple OLEDs were measured at different Liq doping concentrations does not reflect difficulty with identifying the correct concentration for optimizing the OLED configurations; to the contrary, this confirms it was well within the POSITA's skill to identify the optimal Liq concentration in any given OLED stack. Ex. 2003 (Kim, 2011) at 54-55, 60. This paper also shows that a POSITA would have expected success in Liq-doping the ETL in a variety of different OLEDs, because the experimental OLEDs were functional throughout the full ranges of Liq dopant concentrations tested. That different doping concentrations may need to be determined for a given OLED stack to optimize charge balance does not teach away from n-doping with Liq. This is particularly the case here because the claims do not require an OLED to be highly optimized; the claims only require an *operable* OLED.

63. Dr. Pattison points out that Ma emphasizes the importance of maintaining charge balance as a purported reason that a POSITA would not have attempted to use Liq-doped ETL in Diekmann's or Suzuki's OLEDs. Ex. 2018, ¶ 57, Ex. 2002, ¶ 130-131. But in Ma, doping the ETL with Liq is precisely what resulted in a better charge balance. Ex. 1010 (Ma) at Abstract, 812-814; Ex. 1003, ¶ 75, 119, 142-143, 204, 226. A POSITA would thus infer the opposite conclusion—that using Liq as an n-dopant could improve charge balance, and thus have been motivated to use Liq to n-dope ETL, with reasonable expectation of success based on Ma's experimental results.

F. A POSITA would have reasonably expected success in using Liq as the n-dopant in Diekmann and Suzuki's ETLs

64. Dr. Pattison argues that designing an OLED stack with a Liq-doped ETL would have been challenging and required significant experimentation to make work in an iridium-based PHOLED. Ex. 2018, ¶ 62-63. I disagree.

65. By the time of the '425 patent, PHOLEDs with p- and n-doped transport layers were a well-established technology. As noted by Walzer et al., the "p-i-n architecture has been extended to phosphorescent OLEDs." Ex. 2101 (Walzer, 2007) at 1254-1255. The work referenced by Walzer is the work of Pfeiffer et al. from 2002. Ex. 1016 (Pfeiffer, 2002) at 1633. By the time of the '425 patent years later, many others had successfully created PHOLEDs with p- and n-doped transport layers, including for both monochromatic OLEDs, Ex. 2101 (Walzer, 2007) at 1256-1257; Ex. 1019 (He, 2004) at 5773-5774, Fig. 1; Ex. 1029 (Gross, 2005) at 7, 9-19; Ex. 1030 (He, 2004) at 3911-3912, Fig. 1, and white OLEDs with multiple emissive layers, Ex. 2101 (Walzer, 2007) at 1257; Ex. 1053 (Schwartz, 2008) at 053311-1-053311-2, Fig. 1; Ex. 1054 (Schwartz, 2006) at 61920Q-1-61920Q-2, Fig. 1.

66. At the same time, Liq was also a well-studied material. As I have explained above, for example in ¶ 26-31, Liq was a known material and n-dopant, and was known to operate analogously to LiF, a preferred n-dopant for the ETL in Diekmann's PHOLED. Further, identifying the appropriate doping concentration was within the skill of a POSITA. Diekmann and Suzuki had blocking layers, which were

well-known in the art and used effectively in a host of other doped PHOLED systems.

This shows a POSITA would expect success in making the simple modification of Diekmann's and Suzuki's OLEDs to use Liq as the ETL n-dopant. As discussed above, such modifications were routine and well within the skill of a POSITA.

67. Dr. Pattison refers to the “delicate and complicated nature” of the relationship between the layers of the stack and problems with Liq and PHOLEDs as a reason that a POSITA would not have reasonably expected success in using Liq as the n-dopant in Diekmann's or Suzuki's stacks. Ex. 2018, ¶ 58. As already discussed above, OLED stack design followed a modular approach with an expectation of success in replacing known components for certain layers with other known components that serve the same function. At the time of the '425 patent, Liq did not have documented problems that would dissuade its use; to the contrary, it had known benefits. Quenching problems had known solutions. And identifying appropriate doping concentrations for good charge balance was within the skill of a POSITA.

68. Dr. Pattison argues that doping of organic semiconductors was unpredictable. Ex. 2018, ¶ 61, Ex. 2002, ¶ 103. The mere fact that the precise mechanism was not fully understood does not mean doping was unpredictable. The literature discussing the effects of Liq doping shows that OLED researchers were successful in making OLEDs incorporating Liq as the n-dopant, despite not having a complete understanding of Liq's mechanisms. For example, doping was being widely

demonstrated with success in OLEDs for years. Endo and Ma demonstrated Liq-doped ETLs in 2002 and 2008. Ex. 1049 (Endo, 2002) at L800-L802, Fig. 1; Ex. 1010 (Ma) at 810-811. The “p-i-n” architecture has been used in PHOLEDs since as early as 2002. Ex. 2101 (Walzer, 2007) at 1254-1255; Ex. 1016 (Pfeiffer, 2002) at 1633. And work on PHOLEDs having both p- and n-doped transport layers has continued over the years, both monochromatic OLEDs, Ex. 2101 (Walzer, 2007) at 1256-1257; Ex. 1019 (He, 2004) at 5773-5774, Fig. 1; Ex. 1029 (Gross, 2005) at 7, 9-19; Ex. 1030 (He, 2004) at 3911-3912, Fig. 1, and white OLEDs with multiple emissive layers, Ex. 2101 (Walzer, 2007) at 1257; Ex. 1053 (Schwartz, 2008) at 053311-1-053311-2, Fig. 1; Ex. 1054 (Schwartz, 2006) at 61920Q-1-61920Q-2, Fig. 1. Indeed, as Dr. Pattison acknowledges, companies like Novaled were already commercializing doped OLED technology by the time of the '425 patent. Ex. 2101 (Walzer, 2007) at 1256; Ex. 1029 (Gross, 2005) at 7, 9-19; Ex. 2018 at ¶ 22. OLED research routinely involves creating OLEDs without a full understanding of the physical mechanisms. The experiments designed to elucidate those mechanism often come after the fact. Since the functions of the different layers of the OLED stack have been generally well-understood, OLED stack design is modular, and researchers routinely attempt, and succeed in, creating OLEDs with novel materials and architectures that swap out certain materials or layers to create new OLED stacks, without fully characterizing the mechanisms of operation of the new stack *a priori*. For the reasons I have discussed above about the

interchangeability of the n-dopants, moving from using Liq as an ETL n-dopant in Ma's stack to using Liq as the ETL n-dopant in Diekmann's and Suzuki's stacks is a minor leap, and a POSITA would reasonably expect success in making such a modification.

69. Dr. Pattison argues that how quenching and charge imbalance issues caused by Liq would be addressed have not been explained. Ex. 2018, ¶ 65. As explained above, there were well-studied solutions to potential quenching issues in the form of blocking layers, and there is no evidence that there would be any charge balance issues because of incorporating Liq. Diekmann and Suzuki already incorporate blocking layers, and merely adjusting the thicknesses of those layers to optimize the OLED characteristics is again a matter of routine optimization. That Ma has no explicit blocking layer and that a POSITA would not have used BPhen is irrelevant, because Diekmann and Suzuki have the blocking layers, and the proposed modification of those OLEDs to use Liq as the ETL n-dopant does not require BPhen to be incorporated as the ETL in Diekmann and Suzuki's OLEDs. In any case, BPhen has been widely used as the ETL in phosphorescent OLEDs due to its high triplet energy, and frequently as an n-doped ETL. For example, Schwartz et al. have successfully demonstrated white PHOLEDs using multiple emissive layers including an iridium compound, and n-doped BPhen as an ETL. Ex. 1053 (Schwartz, 2008) at 053311-1-053311-2, Fig. 1; Ex. 1054 (Schwartz, 2006) at 61920Q-1-61920Q-2, Fig. 1. Pfeiffer et al. and He et al.

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have also used n-doped BPhen as an ETL with phosphorescent emitters. Ex. 1016 (Pfeiffer, 2002) at 1633-1634, Fig. 1; Ex. 1019 (He, 2004) at 5773-5774, Fig. 1; Ex. 1030 (He, 2004) at 3911-3912, Fig. 1.

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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.

August 11, 2025
Date



Russell J. Holmes, Ph.D.