

Expert Opinion

Provided by Prof. Schanze

I. Qualifications and Professional Background

1. I am currently the Professor and Robert A. Welch Distinguished University Chair in Chemistry at the University of Texas at San Antonio and Deputy Editor of *ACS Central Science*.
2. I earned my B.S. in Chemistry from Florida State University in 1979 and my Ph.D. in Organic Chemistry from the University of North Carolina, Chapel Hill, in 1983. I was a Postdoctoral Research Associate in the Department of Chemistry at the University of North Carolina, Chapel Hill from 1983-1984. I was then a Miller Postdoctoral Fellow in the Chemistry Department of the University of California, Berkeley from 1984-1986. In 1986 I was appointed an Assistant Professor in the Department of Chemistry of the University of Florida. I became an Associate Professor at Florida in 1992 and was appointed full Professor in 1997. From 1997 to 2016 I held various Professorship chairs as outlined in my CV. In 2016 I moved to my current position at the University of Texas, San Antonio, where I serve as the Welch Distinguished Professor.
3. Throughout my career I have earned various honors and awards, including the Globalization Award from American Chemical Society (ACS) in 2018. I was made a Fellow of the ACS in 2011 and won the Florida Award from the Florida Section of ACS in 2009. I have also been awarded many Visiting Professor and Lecturer appointments at various universities throughout Asia. A full list of my Honors and Awards is provided in my CV.
4. I have also held many professional service positions, such as the Chair of the Editor Search Committee for the ACS in 2014 and 2015 and I was the Chair of the ACS Publications Editors Conference in San Diego in 2016. I was Associate Editor of the American Chemical Society (ACS) Journal *Langmuir* from 2000 – 2008. From 2008 - 2023 I served as founding Editor in Chief (EIC) of *ACS Applied Materials & Interfaces*. This is one of the largest technical journals in the field of materials chemistry and materials science. As EIC, I overviewed the review of more than 20,000 papers that are submitted annually for consideration to be published.

5. As a professor of Chemistry, I have an active research interest in optoelectronic devices, including OLED devices and solar cells. Over the course of my career, I have authored many peer-reviewed publications in respected scientific journals within this field. Further details about my academic background and a comprehensive list of my publications can be found on my university profile.
6. All in all, I can look back at more than 30 years of research experience in the field of organic optoelectronic materials and devices. Given my extensive experience and expertise, I am confident that I am qualified to provide an informed expert opinion on the subject matter under discussion.

II. Scope of the Expert Opinion

7. I have been made aware of the pending lawsuit between Pictiva Displays International Ltd. (“Pictiva”) and Samsung Electronics GmbH (“Samsung”) on the basis of the German patent DE 10 2008 056 391 B4; in this context I have been provided with the technical documents in these proceedings, in particular the patent in suit, English translations of Pictiva’s writ of complaint, and Samsung’s answer to the complaint, including an English translation of the expert opinion of Prof. Straßner. I am also aware that the filing date of the patent in suit is Sept. 26, 2008, and that I have to read the patent-in-suit from the viewpoint of the common understanding in the art at that point in time, excluding for that purpose any further developments that may have occurred afterwards. I have also been provided with an English translation of the request that Pictiva wants to assert in the lawsuit. The request is annexed as exhibit 1 to this opinion. My understanding of the patent-in-suit is based on the parallel US patent 8,558,223 B2 which is annexed as exhibit 2.
8. The preparation of my expert opinion is based on my best knowledge. The experiments reported here were conducted under my supervision and were done with all due scientific care. Specifically, I have been asked to:
 1. Study and evaluate the arguments presented by Prof. Straßner in his expert opinion.
 2. Opine on whether lithium 8-quinolinolate (LiQ, see Figure 1, top) can be considered as a metal complex in the sense of the patent-in-suit and on whether compound 6,6'-(naphthalene-1,2-diylbis(4,1-phenylene))bis(2,4-diphenyl-1,3,5-triazine) (“compound 742”), (see Figure 1, bottom) will coordinate with LiQ when jointly deposited by co-evaporation.

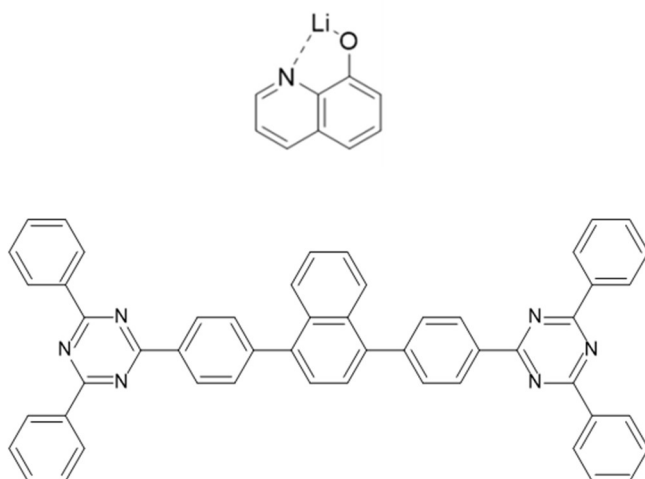


Figure 1: Graphical representation of the molecular structure of LiQ (top) and compound 742 (bottom).

3. To experimentally verify whether compound 742 coordinates to LiQ when jointly deposited under common co-evaporation conditions.
4. To assess, on the basis of my findings above, whether the co-deposition of LiQ and 742 can be expected to provide benefits as described in the patent-in-suit.

III. Summary of my Conclusions

9. Prof. Straßner's expert opinion is concerned with two main topics, namely
 - a) the question whether compound 742 can be expected to have coordinated with LiQ during co-evaporation, and
 - b) the question whether metal complexes other than transition metal complexes such as LiQ are generally compatible to the teaching of the patent-in-suit and, more specifically, whether the coordination of compound 742 with LiQ can be expected to provide the technical benefits mentioned in the patent-in-suit.
10. While I respect Prof. Straßner's views, I must respectfully disagree with his conclusions. I note and have to criticize that Prof. Straßner reached his conclusions without supporting them with first-hand experimental data or demonstrating that his reasoning is based on views accepted in the scientific community.
11. To give one example, Prof. Straßner concluded in paragraph 3(b) of his opinion that LiQ is commonly present as LiQ hexamers which are "*exceptionally stable and a coordinative interaction of the lithium cations with $^{742}\text{C}_{52}\text{H}_{34}\text{N}_6$ is practically*

impossible for energetic reasons.” However, this conclusion is only based on his own assumptions and theories without adequately documenting that these assumptions and theories are scientifically sound and have precedence, for instance by pointing to appropriate statements in peer-reviewed scientific publications. As the former Editor-in-Chief of a very well-respected scientific journal, I have to say that the quality of Prof. Straßner’s opinion is disappointingly low and its results are certainly not fit for publication.

12. Already for this reason, Prof. Straßner’s conclusion that LiQ and compound 742 will not coordinate is not persuasive. However, there are further reasons: His unproven theories ignore a fundamental law of nature, namely the Second Law of Thermodynamics. The scientific literature frequently reports coordination of LiQ with pyridinic nitrogen which is the functional group in compound 742 capable of coordinating with LiQ. Finally, my own experiments demonstrate that Prof. Straßner’s main conclusion, namely that compound 742 will not coordinate with LiQ during co-evaporation, is unfounded.
13. In overall summary, I would strongly expect the co-evaporation of LiQ with compound 742 to result in the coordination of these compounds in an electron transporting layer and I would further strongly expect that this coordination improves the electron conductivity of compound 742 when used as an ETL material.
14. His second conclusion is in my opinion also unfounded: I will explain that there is no technical reason why the metal complex LiQ should not be considered as a metal complex of the patent-in-suit and that LiQ is in my opinion capable of serving the purposes of the metal complex as described in the patent-in-suit. I see this position confirmed by the scientific literature which documents that LiQ is frequently added to triazine-based electron transfer materials (a class of compound to which compound 742 belongs) to improve the electron transfer properties of these materials.

IV. Coordination of Compound 742 to LiQ

15. Prof. Straßner concludes in paragraph 3(b) that LiQ is commonly present as LiQ hexamers which are “*exceptionally stable and a coordinative interaction of the lithium cations with $^{742}\text{C}_{52}\text{H}_{34}\text{N}_6$ is practically impossible for energetic reasons.*”
16. I note that this conclusion is not supported by experiments. This is surprising to me since he should have access to both LiQ and compound 742 via Samsung.

17. Instead, Prof. Straßner builds the following line of argument in paragraphs 5(a) and 5(b) which I summarize here in my own words: During co-evaporation, LiQ is actually present in a hexameric form as Li_6Q_6 . The coordination of compound 742 to the hexameric Li_6Q_6 presupposes that hexameric Li_6Q_6 is first broken apart. This requires a large amount of energy, because the hexameric form is energetically very stable. The energy required for breaking apart hexameric Li_6Q_6 must come from energy released when compound 742 coordinates to LiQ. Since coordinating compound 742 to LiQ releases much less energy than is required to break apart hexameric Li_6Q_6 no reaction between LiQ and compound 742 is taking place.
18. This line of argumentation is not persuasive to me for a number of reasons:
19. First, I note that many of the individual steps of this line of argumentation are not documented with scientific literature. In particular, Prof. Straßner states in paragraph 5(b) that compound 742 is sterically more demanding than quinoline and concludes that this implies that the energy gained from a coordination of compound 742 is far too low to fuel breaking apart hexameric Li_6Q_6 . This approach is questionable since Prof. Straßner exclusively considers steric demand which is only one of many factors determining the feasibility of a chemical reaction (other important ones being e.g. electronic effects and charge distribution).
20. Second, what particularly struck me as wanting is that Prof. Straßner assumes that LiQ is present in the gas phase during the co-evaporation in hexameric form as Li_6Q_6 . I must respectfully disagree with this assumption:
21. In paragraph 5(a), Prof. Straßner points to literature footnote [3] as a first scientific source. Having reviewed the reference (it is attached as exhibit 6), I find no support for such an assumption: This document describes the single-crystal X-ray structure of LiQ, i.e. of LiQ in its solid state. No structural information is provided on how LiQ is present in the gas phase during the co-evaporation. The same is true for the literature footnotes [4] and [5] cited in this context in paragraph 5(a): [4] is attached to my report as exhibit 7 and [5] merely refers back to [3], i.e. is exhibit 6. In footnote [4], i.e. exhibit 7, reference is made to LiQ in the solid state and not how it is present in the gas phase during the co-evaporation.
22. Therefore, despite the reference to literature, Prof. Straßner does not provide tangible scientific support that LiQ is present in the gas phase during the co-evaporation in hexameric form.
23. Prof. Straßner seems to base his conclusions mainly on a study referenced as footnote [6] and attached here as exhibit 4. In this paper, the authors investigated

the structure of various LiQ-based compounds, including neat LiQ, using DFT calculations. Prof. Straßner infers from these DFT calculations that LiQ is unable to react with compound 742 in the gas-phase during co-evaporation. I must respectfully disagree since it ignores a fundamental law of nature, namely the Second Law of Thermodynamics, and, thus, leads to improper results. I will explain this in more detail below:

24. DFT is a quantum mechanical method used to predict the electronic structure of molecules and materials by focusing on electron density rather than individual electron behavior. While DFT is a powerful tool for predicting molecular geometries, it is critical to understand its limitations. DFT calculations provide insights into the ground-state electronic or DFT energies of molecules at 0 K (cf. [exhibit 8](#), page 113). This means they do not account for the effects of temperature or entropy, both of which are fundamental to understanding a system's behavior. Thus, while DFT calculations can offer valuable preliminary information about molecular structures, they cannot predict how these structures will behave under real-world conditions, such as elevated temperatures.

25. In the described study by Rajeswaran *et al.* referenced as footnote [6], average binding energies (ABE) were calculated for various stable LiQ complexes. The ABE is determined by calculating the total energy of the LiQ complex and the energy of the LiQ monomers, then subtracting the latter from the former. More specifically, the energy of the complex is divided by the number of LiQ monomers in the complex, resulting in the electronic energy of the monomer within the complex. From this value, the DFT energy of the individual monomer is subtracted, yielding the average binding energy, which decreases with increasing stability of the complex at its ground state (see Equations (1) and (2)).

$$\begin{aligned} & \text{Energy of monomer in complex} \\ &= \frac{\text{Energy of complex}}{\text{Number of monomers in complex}} \end{aligned} \quad (1)$$

$$\begin{aligned} \text{ABE} &= \text{Energy of monomer in complex} \\ &\quad - \text{Energy of individual monomer} \end{aligned} \quad (2)$$

26. Their findings show that staggered hexameric structures of LiQ exhibit the strongest bonds, leading the authors to conclude that these hexamers are the most stable and thus predominantly observed in experiments.

27. However, it is crucial to reiterate that these calculations are based on the ground-state DFT energy at 0 K, and therefore cannot be directly compared to real-world

conditions. If these DFT values were to be compared to any measurable quantity, they would be most analogous to dissociation enthalpies (ΔH_{diss}).

28. In real-world conditions, the dissociation of complexes follows thermodynamic principles, specifically that dissociation occurs when the Gibbs free energy change (ΔG_{diss}) during the dissociation is negative. According to the second law of thermodynamics, Gibbs free energy is determined from both enthalpy and entropy (cf. [exhibit 9](#), chapter 3.5), as described in the following Equation 3:

$$\Delta G_{diss} = \Delta H_{diss} - T\Delta S_{diss} \quad (3)$$

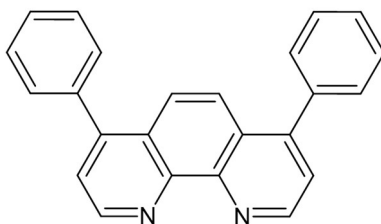
29. In the case of a stable hexameric structure, the bond energy (ΔH_{diss}) is positive, indicating that dissociation does not occur spontaneously at room temperature. However, as temperature increases, the entropy term ($T\Delta S_{diss}$) becomes increasingly more significant. Entropy is a measure of the system's degree of order. Consequently, the dissociation from 1 hexameric LiQ complex to 6 LiQ monomers increases the system's entropy ($\Delta S_{diss} > 0$) and is thus conducive to the dissociation of the hexameric complexes. Multiplied with the temperature, this means that the likelihood of dissociation increases with temperature.
30. Given that very high temperatures are required to vaporize LiQ, even at low pressures ($T > 285^\circ\text{C}$ at 10^{-9} mbar), it is reasonable to assume that the hexameric structure of LiQ will dissociate into smaller clusters or monomers under co-evaporation conditions.
31. This assumption is further supported by Le Chatelier's principle, which suggests that at low absolute pressures, the system will favor the formation of a greater number of molecules (cf. [exhibit 9](#), chapter 7.3), shifting the equilibrium toward dissociation products.
32. Prof. Straßner has not considered the Gibbs free energy relationship and Le Chatelier's principle and, thus, ignored crucial factors following from the Second Law of Thermodynamics. For these reasons, his assumptions are in my opinion critically flawed.
33. Considering the binding energy for hexameric Li_6Q_6 of ~ 238 kcal/mol which is calculated at 0 K in Rajeswaran *et al.*, and combining this with the increase in molecular entropy at elevated temperatures, it is highly likely that Li_6Q_6 hexamers dissociate into smaller clusters or monomers during co-evaporation conditions.

34. In conclusion, Prof. Straßner draws the wrong conclusion from the theoretical structure calculations. The calculations do not account for the significant effects of temperature and entropy. When these factors are considered, it is highly likely that a mixture of LiQ structures, including monomers, exists under the high-temperature and vacuum conditions present during a co-evaporation of LiQ and compound 742.
35. A final remark, it may be that Prof. Straßner may have overinterpreted a passage in Kathirgamanathan *et al.*, i.e. footnote [4] or exhibit 7. In this study, various techniques were employed to analyze the crystal structure of solid LiQ. One such technique was fast atom bombardment-mass spectrometry (FAB-MS), which involves ionizing the solid analyte in a liquid matrix via bombardment with neutral atoms. The ionized component is then transferred into the gas phase for analysis via mass spectrometry.
36. While hexameric clusters were observed in the FAB-MS, it is important to realize that the FAB-MS process is performed at room temperature, i.e. at temperatures more than 250°C lower than given under co-evaporation conditions. At such low temperatures, the aforementioned entropy has a substantially less pronounced influence on the Gibbs free energy. Therefore, the FAB-MS measurement is not representative for what happens under co-evaporation conditions.
37. In summary, I do not see conclusive evidence in the literature to suggest that Li₆Q₆ hexamers persist in the vapor phase under the elevated temperatures described in the patent. Rather, it is highly likely that Li₆Q₆ hexamers dissociate into smaller more reactive clusters or monomers during co-evaporation conditions.
38. The smaller more reactive clusters or monomers of LiQ which are formed during co-evaporation can also be expected to form coordination compounds with compound 742. Compound 742 is comprising six pyridinic nitrogen atoms, i.e. nitrogen atoms which are part of an aromatic ring system. The patent-in-suit describes in the section discussing the appropriate combination of metal complexes and organic compounds that these pyridinic nitrogen atoms are excellent choices in coordinating to metal complexes. See exhibit 2, col. 11, ll. 17-52, which I am told corresponds to paragr. [0034] of the patent-in-suit. The excellent ability of pyridinic nitrogen atoms to coordinate to metal complexes is also not limited to specific compounds but generally given for the entire class of compounds. This is not only evident from the large structural diversity of the preferred compounds (I) to (XXIII) of the patent-in-suit but also further supported by the scientific literature which reports coordination of LiQ with the pyridinic nitrogen atoms:

39. Kang *et al.* report in exhibit 11 on page 140207, right col:

“the complexation of ETL (BPhen) and Liq was proved by analyses of the Gibbs – DiMarzio prediction and X-ray photoelectron spectroscopy[16].”

40. As a side remark, for the benefit of the Court, the reference to complexation in the above quote means the same as coordination in the sense of the patent-in-suit, i.e. reference is made to a coordination of LiQ with the pyridinic nitrogen atoms of BPhen. For further information, BPhen has the following structure:

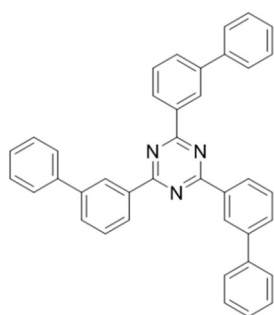


41. In the above quote, Kang *et al.* reference lit. [16] which is attached to my opinion as exhibit 13. In exhibit 13, a film obtained by co-evaporation of LiQ with BPhen is analyzed by X-ray photoelectron spectroscopy (XPS). It is reported in the paragraph bridging the columns of page 1678 and the following paragraph that the XPS signals of the pyridinic nitrogen and of the lithium are synchronously shifted and that this implies a chemical interaction between LiQ and the pyridinic nitrogen atoms of BPhen and not just the mere co-existence “side-by side” as it is present in a purely physical mixture:

“X-ray photoelectron spectroscopy (XPS) was performed to investigate molecular interaction between BPhen and Liq. ... In case of BPhen, the N 1s peak of pristine BPhen sample was centered at 398.2 eV, which could be assigned to sp²-hybridized pyridinic nitrogen in the BPhen molecule. Interestingly, the N 1s spectra of the mixture films cannot be fully deconvoluted with the N 1s spectra of pristine Liq and BPhen (see Figure S4 in the Supporting Information), implying that Liq and BPhen are not simply in their physical mixture, but their chemical interaction exists in the mixture. In the Li 1s spectra, 100% Liq showed a peak at 55.6 eV. When 17% of Bphen was mixed to Liq, a red-shift of Li 1s peak to 55.1 eV was observed and no additional shifts were observed as the molar ratio of Bphen increased further. The synchronous peak shifts observed in N 1s and Li 1s spectra upon mixing Bphen and Liq, the binding energy increase in N 1s from pristine Bphen (~0.4 eV) and the binding energy decrease in Li 1s from 100% Liq (~0.5 eV), imply some chemical interaction between the N atoms (Bphen) and the Li atom (Liq).”

42. Thus, there is direct experimental evidence of the pyridinic nitrogen atoms of BPhen coordinating to the LiQ metal complex.

43. Moreover, [exhibit 11](#) demonstrates by way of DFT calculations how further compounds containing pyridinic nitrogen atoms coordinate to LiQ. These compounds are T2T, i.e. a triazine derivative like compound 742, and BPyTP2 which contains 4 pyridine rings:

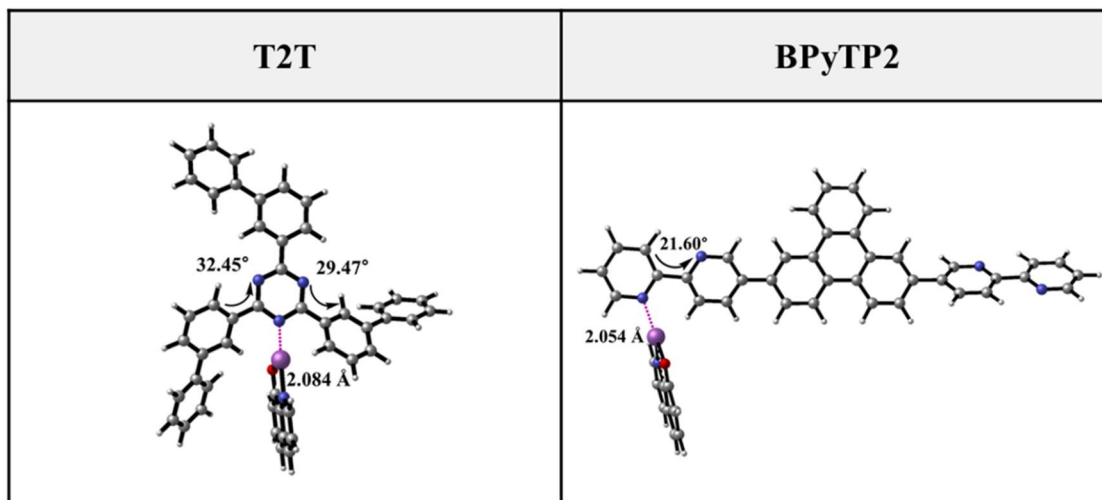


T2T



BPyTP2

44. The following structures for the coordination compounds are reported:



45. As can be seen in the above structures, blue pyridinic nitrogen atoms coordinate with the magenta-colored lithium atom. The coordination is shown as a dotted pink line.

46. To sum up, the smaller more reactive clusters or monomers of LiQ which are formed during co-evaporation can also be expected to form coordination compounds with compound 742 since the pyridinic nitrogen atoms are excellent

coordination formers with LiQ irrespective of the exact chemical structure of the compound. The latter is supported by the patent-in-suit which cites many structurally very diverse compounds comprising pyridinic nitrogens as preferred compounds and the scientific literature which demonstrates coordination via pyridinic nitrogen atoms of equally diverse compounds.

47. I was further tasked with conducting a series of experiments to determine whether a coordination complex forms when LiQ and a compound 742 are co-evaporated. Unfortunately, compound 742 is not commercially available. We therefore opted to use a model compound that is very similar in structure and chemical behavior to compound 742. We used 4,4'-bis(4,6-diphenyl-1,3,5-triazin-2-yl)biphenyl (BTB) which has a very similar structure and in particular shares the diphenyltriazine moieties of compound 742:

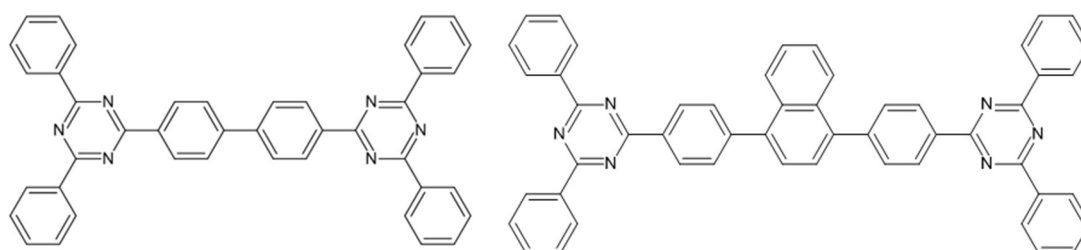


Figure 2: Comparison of the molecular structure of BTB (left) and compound 742 (right). Both compounds are structurally related, differing solely by the naphthalene moiety in the middle.

48. As said, BTB was selected due to its structural and chemical similarity to compound 742, making it an appropriate model compound for our investigation. BTB is an ideal substitute for compound 742 as it retains the critical structural motifs necessary to assess the coordination behavior. I have no doubts that BTB's ability to coordinate to LiQ under co-evaporation conditions will be equivalent to that of compound 742. Therefore, the results obtained for BTB are in my view representative for compound 742.
49. We have deposited LiQ and BTB by co-evaporation at estimated molar ratios of 30:70 and 50:50, in addition to evaluating the neat compounds BTB and LiQ for reference purposes. All 4 deposited films were analyzed using X-ray photoelectron spectroscopy (XPS). XPS is a surface-sensitive technique used to study the elemental composition and chemical states of a material's surface. It works by irradiating the sample with X-rays, which eject electrons from the atoms. The kinetic energy of these electrons is measured, which reveals information about the binding strength of the electrons and thereby the present elements as well as their chemical environment. XPS is widely used for molecular structural analysis.

50. The experimental parameters for both the co-evaporation/film preparation and the XPS measurements were as follows:

- Film deposition:

- Chemicals:

AmScope Microscope Slides with a length and width of 1 in and a thickness of 0.8-1.0 mm were used as glass substrates onto which the BTB/LiQ films were prepared, sublimed BTB with a purity of > 99 % (LCMS) was sourced from Osilla, and sublimed LiQ with HPLC grade purity (> 99.5 %) was bought from Sigma Aldrich.

- 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.

- Deposition:

Glass substrates were first cleaned by ultra sonication for 15 minutes each, sequentially in a sodium dodecyl sulphate solution, isopropanol, and acetone. The substrates were dried under nitrogen for 30 minutes.

- a) neat BTB 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 BTB was placed in a crucible and the chamber was closed and evacuated to a pressure of 10^{-9} mbar. A rate of $1 \text{ \AA/s}$ is used to evaporate BTB at $\sim 280^\circ\text{C}$ and achieve a film thickness of $\sim 200 \text{ nm}$.

- b) neat LiQ 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 was placed in a crucible and the chamber was closed and evacuated to a pressure of 10^{-9} mbar. A rate of $1 \text{ \AA/s}$ was used to evaporate LiQ at $\sim 310^\circ\text{C}$ and achieve a film thickness of $\sim 200 \text{ nm}$.

- c) BTB-LiQ co-deposition

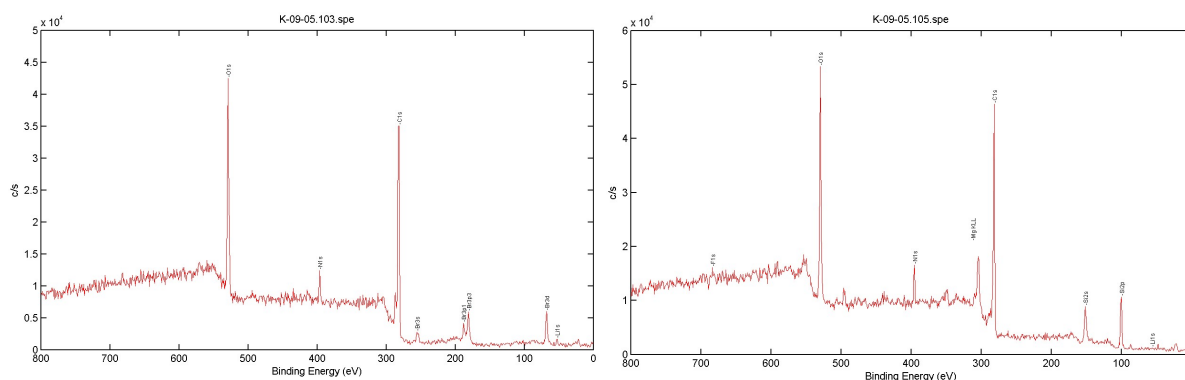
4 cleaned and dried glass substrates were loaded on a substrate holder and mounted in the vacuum evaporator for film deposition. In the evaporating chamber, 50 mg of LiQ and 50 mg of BTB were placed in two separate crucibles with separate heating sources and the chamber was closed and evacuated to a 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 ~ 290 °C and LiQ at ~ 300 °C and achieve a film thickness of ~ 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 ~ 280 °C and LiQ at ~ 310 °C and achieve a film thickness of ~ 200 nm.

- XPS measurements:

XPS measurements were conducted externally at Rice University in Houston, Texas, USA, using a PHI Quantera XPS, which uses a focused monochromatic Al K α X-ray (1486.7 eV) source for excitation. The 50 W, 15 kV and 200 μ m diameter X-ray beam was pointed at the sample. XPS scan spectra in the 800 - 0 eV binding energy range were recorded in steps of 0.5 eV with a pass energy of 224 eV. High resolution scan spectra were recorded in steps of 0.1 eV with a pass energy of 26 eV. Low energy electrons and Ar ions were applied for specimen neutralization in each measurement. All peaks were calibrated with a C 1s position of 248.8 eV. A take-off angle of 45 degree was used in all measurements, with a sampling depth of approximately 3-5 nm.

51. In the following, I provide my evaluation of the obtained data:

52. Figure 3 shows the raw XPS spectra, with the top row of Figure 3 displaying the data for neat LiQ and BTB, respectively, and the bottom row of Figure 3 presenting the results for the co-evaporated films with LiQ:BTB ratios of 30:70 and 50:50 respectively.



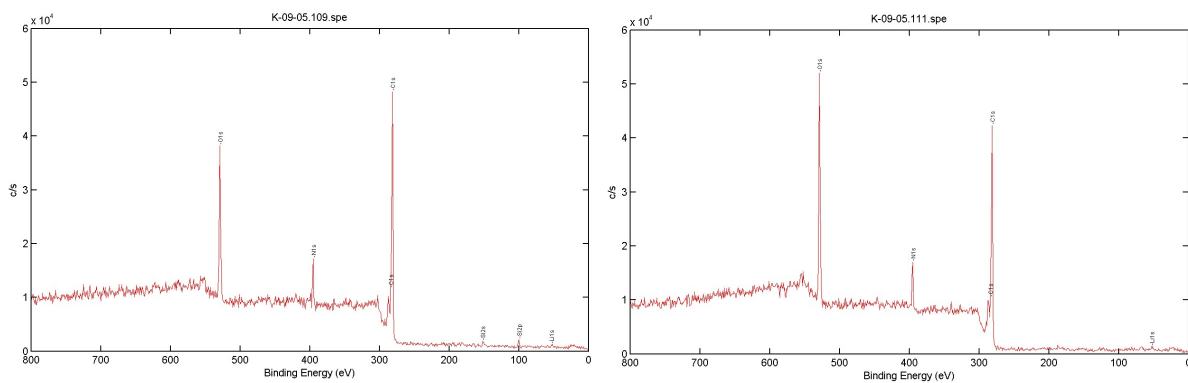


Figure 3: Raw sequence scans of the 4 prepared samples. Distinct binding energy bands are observed. Those graphs are the basis for the analysis shown in Figure.

53. Distinct binding energy bands are observed, and the Li 1s binding energy is of particular interest. Although the Li signal is inherently weaker than those of other elements, it remains clearly distinguishable from the underlying noise. Figure 4 presents the background-corrected Li 1s binding energy distributions for all samples, where the x-axis shows the binding energy in electron volts and the y-axis shows the intensity of the signal at a given binding energy.

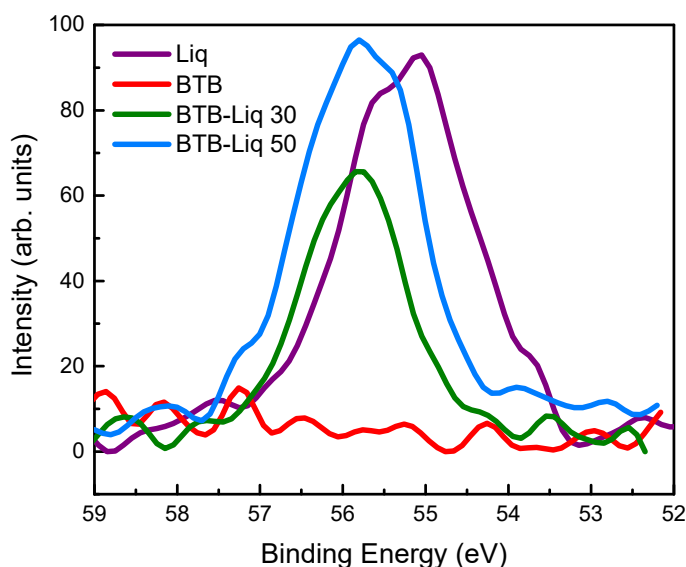


Figure 4: Background-corrected Li 1s binding energy distributions for the prepared films measured via XPS. The x-axis shows the binding energy in electron volts, while the y-axis shows the corresponding intensities at a given binding energy.

54. As expected, the neat BTB sample shows no Li 1s signal, as it contains no Li. In contrast, clear Li 1s signals are evident in the neat LiQ sample and the co-evaporated films. A notable shift in the Li 1s binding energy by approximately 1 eV to higher values is observed when comparing the neat LiQ sample to the co-evaporated films. This shift indicates a substantial change in the electronic environment surrounding the Li ion. The observed shift towards higher binding

energies suggests a reduction in electron density around the Li ion, cf. exhibit 13, page 1678, right col., 2nd paragr.

55. This finding can be interpreted as follows: In LiQ, the coordination bond of the quinolinolate anion (i.e. Q in LiQ) transfers electron density to the lithium cation. In other words, the coordination of the quinolinolate anion with the lithium cation creates a metal complex which has an increased electron density at the lithium cation. Once BTB coordinates to the lithium cation of LiQ, a part of the complex's electron density is further transferred to the ligand BTB. This is a plausible assumption since triazine derivatives such as BTB are known to be electron-deficient aromatics (cf. exhibit 10, page 2, 1st paragr.), i.e. have an affinity for more electron density. In other words, there is a metal complex-to-ligand charge transfer taking place which explains the shift of the lithium signal.

56. In any case, the XPS results match those of exhibit 13 (discussed above) in that the results demonstrate that there is an interaction on the chemical level which goes beyond the mere coexistence of LiQ and BTB as a physical mixture.

57. In conclusion, these experimental results provide compelling experimental evidence for the formation of a coordination bond between LiQ and BTB.

58. Comprehensively considering my above findings based on the scientific literature and my experimental results for the model compound BTB, I have no reasonable scientific doubt that the closely related compound 742 will coordinate with LiQ under co-evaporation conditions.

V. LiQ is a metal complex in the sense of patent-in-suit and provides the benefits mentioned in the patent

59. Prof. Straßner further addresses in paragraphs 5(c) and 5(d) whether metal complexes other than transition metal complexes such as LiQ are generally compatible to the teaching of the patent-in-suit and, more specifically, whether the coordination of compound 742 with LiQ can be expected to provide the technical benefits mentioned in the patent-in-suit.

60. From my above explanations, it is already evident that LiQ is a metal complex which coordinates well with pyridinic nitrogen atoms which are contained in almost all of the preferred compounds (I) to (XXIII) of the patent-in-suit. That lithium complexes such as LiQ are able to coordinate with up to 8 ligands was well-known at the priority date of the patent-in-suit, cf. exhibit 4, published in 2007, stating in the introduction:

*“Lithium 8-hydroxyquinolate (LiQ) is of current interest in the organic light-emitting diode (OLED) industry as a host material in emission layers [1,2] and as interface materials for the electron-transporting and electron-injecting layers for OLED devices, [2–5]. In recent years, the crystal structure of complexes containing lithium has received increased attention. **Coordination numbers in lithium complexes are known to vary and occur in the range of 2–8, [6,7], with smaller numbers more common.**”*

61. Thus, I have no doubt that the skilled reader of the patent-in-suit is aware of the fact that lithium ions are excellent in coordinating with a plurality of ligands. I also have no doubt that the skilled reader would consider that the pyridinic nitrogen atoms present in many of the patent’s preferred examples will readily coordinate to lithium complexes such as LiQ. I reach this conclusion not the least because the patent-in-suit describes in the section discussing the appropriate combination of metal complexes and organic compounds that pyridinic nitrogen atoms are excellent choices for ligands coordinating to metal complexes. See exhibit 2, col. 11, ll. 17-52, which I am told corresponds to paragr. [0034] of the patent-in-suit.
62. In conclusion, I find it not persuasive that Prof. Straßner doubts that the skilled reader would consider lithium complexes as metal complexes which are suitable for the purposes of the patent-in-suit.
63. Prof. Straßner further questions in paragraphs 5(c) and 5(d) whether the coordination of compound 742 with LiQ can be expected to provide the technical benefits mentioned in the patent-in-suit. I have to respectfully disagree:
64. As already explained, the patent-in-suit states that in order to significantly improve electron conductivity within the electron transport layer, the metal complexes must be able to transfer electrons from the metal complex to the ligand.
65. Moreover, as also already explained, the XPS results of my experiments with BTB can be interpreted as a metal complex-to-ligand charge transfer. I have no doubt that the very similar compound 742 will also show the same XPS trend that can be explained as metal complex-to-ligand charge transfer event.
66. Still further, the DFT calculations in exhibit 11 further demonstrate the general ability of triazine LiQ coordination compounds to participate in an electron transfer from the complex LiQ to the ligand triazine. The study shows that the highest occupied molecular orbital (HOMO) is entirely located on the LiQ molecule, indicating its role as the electron donor, while the lowest unoccupied

molecular orbital (LUMO) is situated exclusively on the triazine ligand, identifying it as the electron acceptor. See Figure 4 of exhibit 11:

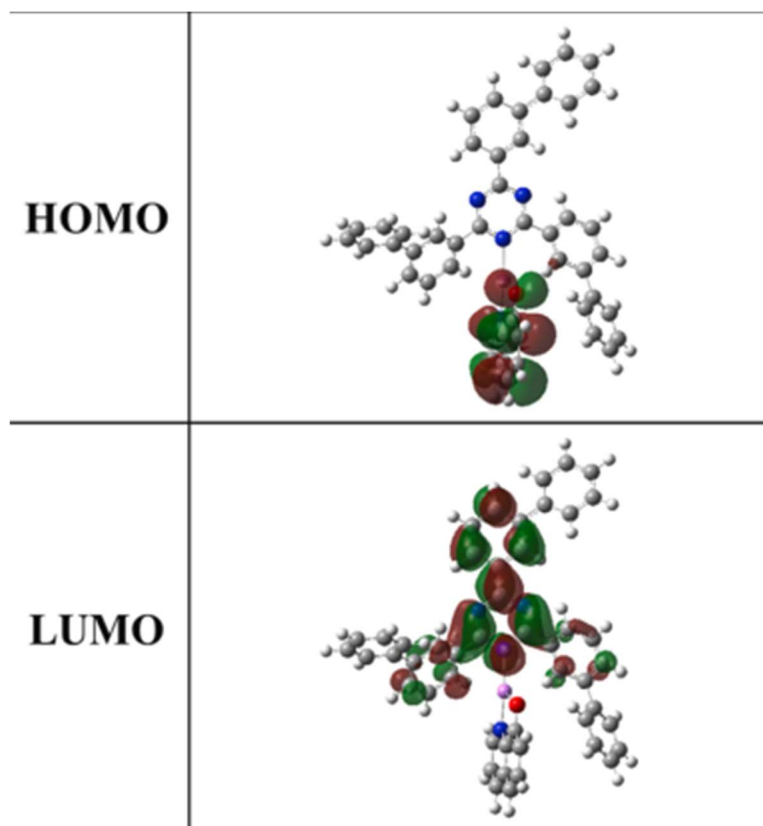


Figure 4 of exhibit 11: HOMO and LUMO states within a organometallic complex formed between LiQ and the organic triazin ‘T2T’. The HOMO is exclusively on the LiQ molecule, while the LUMO is concentrated on the triazine ring (best identified by its 3 blue pyridinic nitrogen atoms in the HOMO).

67. I therefore conclude that the scientific evidence, both experimental and theoretical, suggests LiQ and compound 742 will likely participate in a metal complex-to-ligand charge transfer in an electron transport layer.

68. I see the above conclusion particularly supported by the well-established fact that combining LiQ with electron-deficient aromatic materials, and particularly triazine derivatives, is improving electron mobility within the electron transport layer. For instance, the textbook “*Advanced Display Technology*”, attached in excerpts an exhibit 14, states in the paragraph bridging pages 11 and 12:

“More recently, several designs of electron-deficient molecules with large aromatic planes such as 2,4,6-tris(biphenyl-3-yl)-1,3,5-triazine (T2T)[...] have demonstrated very high mobilities and improved device performance. In addition to selecting materials with high inherent mobilities, the ETL can be

doped to further improve the mobility within the layer. One common class of material for this are organolithium complexes such as Liq”.

69. This shows that LiQ is nowadays an established way of improving electron mobility in the electron transfer layer, which in my opinion is only possible if a coordination between the LiQ molecule and the pyridinic nitrogen of the triazine (like compound 742) exists to transport the electrons.
70. In contrast, I find Prof. Straßner’s argumentation that LiQ cannot provide the technical benefits of the patent-in-suit to not be persuasive. He argues that the Li atom, given its position in the periodic table, has an electron configuration of $1s^2, 2s^1$. Since Li exists as a cation in LiQ, the 2s valence shell is effectively vacant, making lithium cations in his opinion incapable of donating electrons to compound 742. While it is correct that a naked Li ion contains an empty 2s valence shell, this argument overlooks the crucial role of the quinolinolate anion of LiQ, which – as explained – donates electron density to the Li cation and, once coordinated to compound 742, to the electron-deficient aromatic system of compound 742.
71. Thus, in overall summary, I would strongly expect the co-evaporation of LiQ with compound 742 results in the coordination of these compounds in an electron transporting layer and I would further strongly expect that this coordination improves the electron conductivity of compound 742 when used as an ETL material. Moreover, there is no technical reason why the metal complex LiQ should not be considered as a metal complex of the patent-in-suit since it capable of serving the purposes of the metal complex as described in the patent-in-suit. I see this position confirmed by the scientific literature which documents that LiQ is frequently added to triazine-based electron transfer materials to improve the electron transfer properties of these materials, i.e. added to a class of compounds to which compound 742 belongs.

September 16, 2024

K. S. Schanze / 09.16.2024

Prof. Kirk Schanze