Quantum dot light emitting devices for photomedical applications

Hao Chen (SID Student Member)
Juan He (SID Student Member)
Raymond Lanzafame
Istvan Stadler
Hamid El Hamidi
Hui Liu
Jonathan Celli
Michael R. Hamblin
Yingying Huang
Emily Oakley
Gal Shafirstein
Ho-Kyoon Chung (SID Fellow)
Shin-Tson Wu (SID Fellow)
Yajie Dong (SID Member)

***Abstract* —** While OLEDs have struggled to find a niche lighting application that can fully take advantage of their unique form factors as thin, flexible, lightweight and uniformly large-area luminaire, photomedical researchers have been in search of low-cost, effective illumination devices with such form factors that could facilitate widespread clinical applications of photodynamic therapy (PDT) or photobiomodulation (PBM). Although existing OLEDs with either fluorescent or phosphorescent emitters cannot achieve the required high power density at the right wavelength windows for photomedicine, the recently developed ultrabright and efficient deep red quantum dot light emitting devices (QLEDs) can nicely fit into this niche. Here, we report for the first time the in-vitro study to demonstrate that this QLED-based photomedical approach could increase cell metabolism over control systems for PBM and kill cancerous cells efficiently for PDT. The perspective of developing wavelength-specific, flexible QLEDs for two critical photomedical fields (wound repair and cancer treatment) will be presented with their potential impacts summarized. The work promises to generate flexible QLED-based light sources that could enable the widespread use and clinical acceptance of photomedical strategies including PDT and PBM.

***Keywords* —** *quantum dot light emitting devices (QLEDs), photomedicine, photodynamic therapy, photobiomodulation.*

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1 Background for quantum dot light emitting devices and photomedicine

Photomedicine is an emerging medical field, in which light is used either to kill cancer cells with assistance of photosensitizers and singlet oxygen (photodynamic therapy, PDT) or to stimulate cellular function leading to beneficial clinical effects (photobiomodulation, PBM). They have been demonstrated as minimally invasive treatment strategies with expensive, bulky light sources, such as laser or LED arrays.[1] However, photomedicine still has not received widespread clinical acceptance mainly because of the lack of effective, low-cost illumination devices.

Because of their unique form factors as thin, flexible, lightweight and uniformly large-area light sources, organic light emitting devices (OLEDs) were once proposed to work as light-emitting bandages for PDT[2] but were later abandoned in favor of LEDs[3] because photomedical applications generally require light sources of relatively high brightness (>20 000 Cd/m^2 or ~10 mW/cm^2) at wavelengths in the deep red region in order to have deep tissue penetration while still maintaining sufficient energy for molecular excitations.[1] Existing OLEDs with either fluorescent or phosphorescent emitters cannot achieve such high brightness at the right wavelength windows because of significant efficiency roll-off problems of OLEDs at high current density[4] and the lack of efficient deep red emitters with narrow spectra.[5]

Thanks to their size-controlled tunable emission wavelength, narrow emission spectra and simple solution processability, quantum dot light emitting devices (QLEDs) have attracted much attention recently as a candidate for next generation display.[6] Among QLEDs of various colors, red QLEDs are currently most advanced and have demonstrated efficiency and brightness that rival or beat state-of-the-art thermal-evaporated red OLEDs, with narrow peak linewidths in the 20 to 30-nm range. In particular, our group has reported ultrabright and efficient deep red QLEDs.[7] As shown in Fig. 1, the devices demonstrate peak emission wavelength of 620 nm, narrow bandwidth of 22 nm and can

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Hao Chen, Juan He, Shin-Tson Wu and Yajie Dong are with the College of Optics and Photonics, University of Central Florida, Orlando, FL, USA.
Hao Chen and Yajie Dong are with the Nanoscience Technology Center, University of Central Florida, Orlando, FL, USA; telephone 407-823-5159, fax 407-882-2819; e-mail: yajie.dong@ucf.edu.
Raymond Lanzafame is with Raymond J Lanzafame MD PLLC, Rochester, NY, USA.
Raymond Lanzafame and Istvan Stadler are with the Laser Surgical Research Laboratory, Rochester General Hospital, Rochester, NY, USA.
Hamid El Hamidi, Hui Liu and Jonathan Celli are with the Department of Physics, University of Massachusetts Boston, Boston, MA, USA.
Michael R. Hamblin and Yingying Huang are with the Harvard Medical School, Wellman Center for Photomedicine, Boston, MA, USA.
Emily Oakley and Gal Shafirstein are with the Roswell Park Cancer Institute, Photodynamic Therapy Center, Buffalo, NY, USA.
Ho-Kyoon Chung is with the ITRC AMOLED Research Center, Sungkyunkwan University, Korea.
Yajie Dong is with the Department of Materials Science and Engineering, University of Central Florida, Orlando, FL, USA.

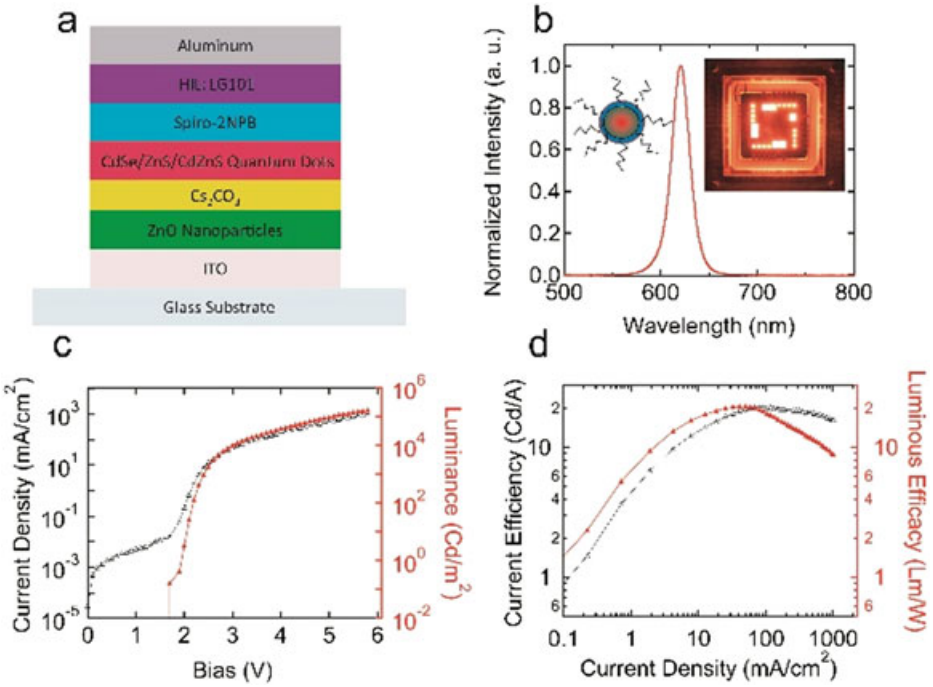

FIGURE 1 — Ultrabright highly efficient, low roll-off inverted quantum-dot light emitting devices (QLEDs). (a) A schematic representation the ultra-bright, high efficiency, inverted QLEDs. (b) Spectra of QLED electroluminescence. (c) Luminance and current density versus driving voltage and (d) luminous efficacy and current efficiency versus driving current density for typical devices. Adapted from data in reference.[7]

achieve high current efficiency (20.5 Cd/A at ~20 000 Cd/m^2) and small efficiency roll-off at high driving current density. Ultrahigh brightness of 165 000 Cd/m^2 can be achieved at current density of 1000 mA/cm^2, which sets a new brightness record for existing organic related red light emitting devices.

Although the relatively short lifetime of these ultrabright deep red QLED devices has limited their immediate applications in display or daily lighting markets, these ultrabright deep red QLED devices can be promising light sources for photomedicine where low-cost, wearable, disposable light emitting bandage products are highly desired. And the narrow emission band and wavelength tunability of QLEDs make it feasible to better fit the emission spectrum into the absorption window of photosensitizers (for PDT) or cytochrome C (for PBM).

In this paper, we present preliminary PBM and PDT results using these ultrabright red QLEDs as excitation light sources, with parallel studies using inorganic LEDs as comparisons. The perspective of tuning QLED wavelength for targeted photomedicine, development of flexible QLED and their potential impact to wound repair or cancer treatment will also be discussed.

2 Preliminary experimental results

A 4-pixel (4 × 4 mm each.) QLED array has been developed as photomedical light source. As shown in Fig. 2, a specialized platform and cradle was built to stabilize the QLED array allowing proper tray positioning underneath cell cultures for in-vitro PBM and PDT experiments. The results are compared with control cell cultures that received no light treatment or parallel studies with inorganic LED treatment.

2.1 Preliminary photobiomodulation results

For PBM testing, three cell lines (HEp-2 [ATCC-CCL-2]; L929 [ATCC-CCL-1], 3T3 [ATCC-CRC-2593]; American Tissue and Cell Culture Collection [ATCC], Manassas, VA), were cultured in 24-well trays in complete Dulbecco's Modified Eagle Medium supplemented with 10% fetal bovine serum, streptomycin–penicillin–fungicin, glutamine and pyruvate, without phenol red. These cell lines are frequently used for in-vitro studies of PBM and are often used as surrogates for whole animal studies of wound healing, among other applications. HEp-2 cells are a human epithelial cell line derived from cervical cancer and are biologically identical to the famous HeLa cell line. The L929 is a mouse fibroblast cell line, and 3T3 cells are also a fibroblast cell type derived from mouse embryos.

Photoradiation was performed using the inverted ultrabright QLED to deliver 4.0 J/cm^2 to the culture wells during 10-min treatment at power density of ~8 mW/cm^2. Control cell cultures received no light treatment. Cell metabolism was assessed by 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide assay (Chemicon International Inc., Temecula, CA) 24 h post treatment. The

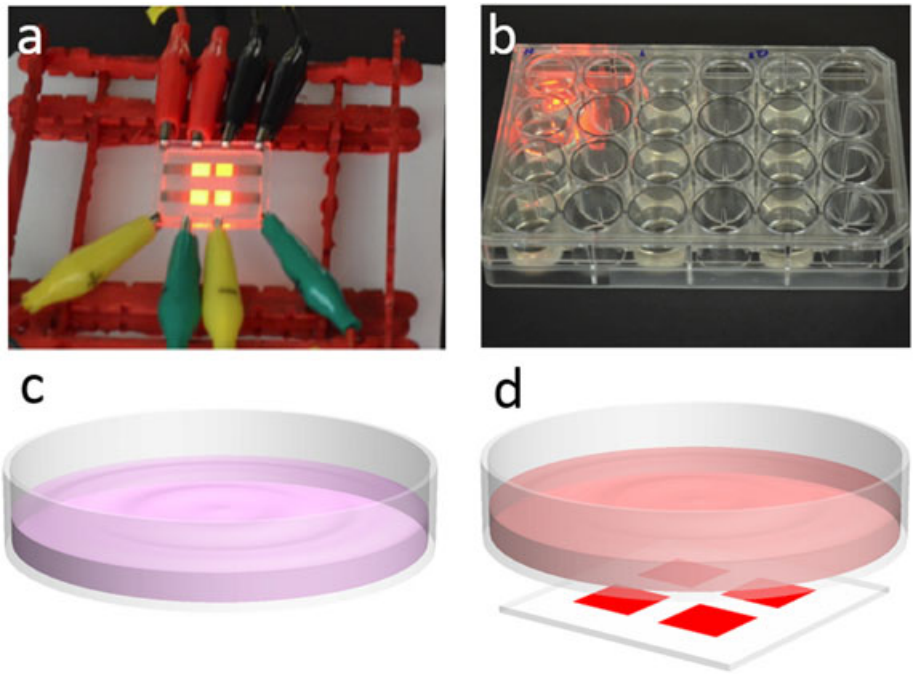

FIGURE 2 — Experimental setup for photomedical testing. (a) 2 × 2 red QLED array; (b) experimental setup; (c) control cell cultures without light treatment; (d) cell cultures using QLED as lighting source.

3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide assay is a colorimetric assay and is a popular method to evaluate cell metabolic activity. Parallel studies were performed at 670 nm ± 20 nm using an LED device (Quantum Devices, Barneveld, WI) delivering 4.0 J/cm^2 during 10-min treatment for comparison with QLED PBM.

Assay results at 24 h (presented in Table 1) show that QLED PBM increased cell metabolism in the HEp-2 cell line and in a similar fashion to a NASA LED source.[8] For HEp-2, L929 and 3T3, QLED PBM increases the cell metabolism by 27.9, 26 and 12.5% over the control systems, respectively. Although the peak wavelength of QLED (~620 nm) is still away from the favorite 660 nm for PBM, the results of QLED BPM are comparable with the LED PBM. Further tuning the emission wavelength of QLED is expected to lead to improved PBM results.

2.2 Preliminary photodynamic therapy results

To evaluate the potential of red QLEDs as a light source for PDT, we used 3D cultures of A431 cells (a human cell line frequently used in cancer-associated biomedical study) grown on beds of laminin-rich-extracellular matrix and photosensitized by administration of aminolevulinic acid (ALA) leading to accumulation of protoporphyrin IX (PpIX) prior to light activation.

Cultures were treated using either the QLED sources with low average irradiance (approx 1.8 mW/cm^2) or a solid state LED with similar spectral emission but higher irradiance (approx 130 mW/cm^2). Dosimetry was controlled so that cultures received the same total light dose of 30 J/cm^2 over the course of either 4.75 h (QLED) or 4 min (solid state

TABLE 1 — 3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide (MTT) assay results at 24 h.

	Mean MTT values		
Light source	HEp-2	L929	3 T3
QLED PBM	0.697 ± 0.082 (N = 14) ††	0.574 ± 0.062 (N = 14)	0.443 ± 0.182 (N = 12)
QLED control	0.545 ± 0.066 (N = 14)	0.510 ± 0.062 (N = 14)	0.351 ± 0.090 (N = 12)
LED PBM	0.789 ± 0.032 (N = 4) **	0.647 ± 0.021 (N = 4) †	0.346 ± 0.036 (N = 4) *
LED control	0.668 ± 0.033 (N = 4)	0.499 ± 0.033 (N = 4)	0.273 ± 0.050 (N = 4)

$^{*}p$ = 0.05.
$^{**}p$ = 0.002.
$^{\dagger}p$ = 0.0003.
$^{\dagger\dagger}p$ = 0.0004 versus control.
Note: p is a value used to assess if the experimental outcome is distinct from the control. Smaller p value (<0.05) indicates significant difference.

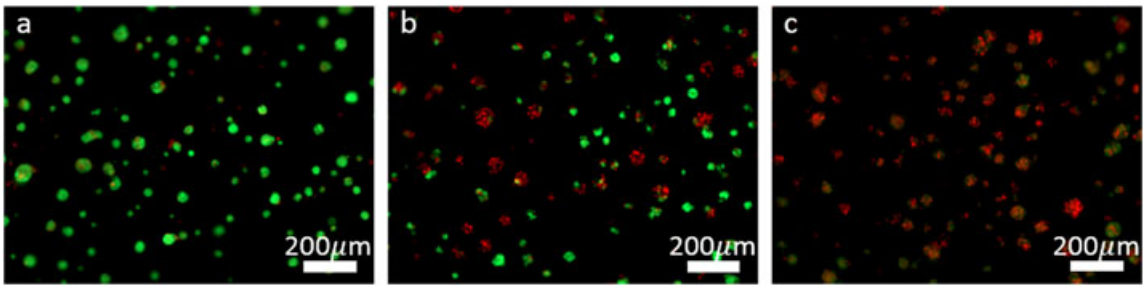

FIGURE 3 — Fluorescent vital-dye labeled 3D cultures 24 h post photodynamic therapy (PDT) treatment. Calcein labels live cells green while ethidium bromide labels dead cells red. (a) Control cells without light treatment; (b) LED-based PDT; (c) QLED-based PDT.

LED). In the case of QLED irradiations, calibration runs were first performed to measure power drop off during extended continuous operation. Irradiation duration was determined by integration of irradiance over time to determine the time required (4.75 h) for the QLED to deliver the same total light dose as the solid state LED source (30 J/cm^2). Treatment response was evaluated 24 h after PDT using an imaging-based approach described previously.[9]

As shown in Fig. 3, both QLED and LED sources achieve photodestruction of 3D tumor nodules, while quantitative image processing of multiple replicates reveals PDT efficacy is slightly enhanced using the QLED source, with residual tumor viabilities of 0.61 +/− 0.04 versus 0.53 +/− 0.08 for the solid state and QLED sources respectively. This result is consistent with previous reports that PDT at low dose rates may be more effective and is significant here in view of the capability of the QLED to act as a low-cost, effective and ergonomic source for PDT light activation over extended periods.

These in-vitro studies are the first to demonstrate PBM and PDT using a QLED device and pave the way for further developments of QLED-based photomedicine.

3 Perspective

3.1 Wavelength tunable red QLED for targeted photomedicine

It should be noted that these promising preliminary results were obtained with ultrabright red QLED with a peak wavelength of ~620 nm. While this wavelength falls into the favorite range for most photomedical applications (620–670 nm), highly effective phototherapy calls for better wavelength specific spectral control to maximize the absorption for photosensitizers (for PDT) or cytochrome C (for PBM) from QLED. By tuning the synthesis conditions (QDs size and composition), we can achieve ultrabright QLEDs with precisely controlled emission peaks at the following wavelengths for wound repair and cancer treatment applications (shown in Fig. 4):

1) 630 nm for porfimer sodium (Photofrin®; PF), a Food and Drug Administration approved photosensitizer widely used for various PDT cancer treatments; 2) 635 nm for PpIX, which is an endogenous photosensitizer that accumulates after administration of ALA and has been developed for a wide range of applications and Food and Drug Administration approved for PDT treatment of actinic keratosis; 3) 660–670 nm for using 2-1 [hexyloxyethyl]-2-devinylpyropheophorbide-A, Chlorin e6 or talaporfin in PDT, and cytochrome C, the primary light absorbing chromophore for PBM.

Currently, such precise wavelength control can only be realized by expensive, bulky lasers, although the laser light needs to be waveguided with optical fibers and spread out with diffusers for large-area applications. Comparing to lasers, QLED has clear advantages as low-cost, large-area and wearable light sources.

3.2 Flexible QLED development

Flexible QLEDs are in obligatory demand to achieve a solution that is functional for a wearable bandage light source. While many groups have demonstrated their flexible devices by simply transferring the QLEDs from rigid substrates to flexible substrates,[10] much progress is still in need to further improve the device performance, reliability and to lower the manufacturing cost:

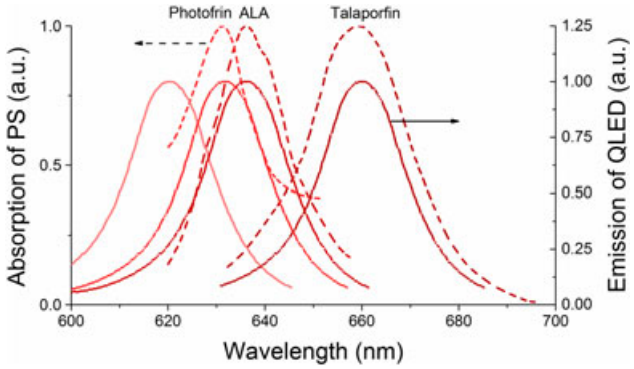

FIGURE 4 — The absorption spectra (dashed line) of some common photosensitizers (PS) and the target emission spectra of QLEDs (solid line) which are slightly shifted from demonstrated 620 nm. Porfimer sodium (Photofrin®), aminolevulinic acid (ALA) and talaporfin are three photosensitizers widely used for various PDT cancer treatments.

1) Transparent conductor material and process development: conventional QLED on rigid substrates uses indium tin oxide film as bottom conductor layer, which is not stable during repeated bending. Ag nanowire transparent conductor structure with superior surface planarity is more suitable for flexible QLEDs; 2) large-area printing of quantum dot for QLED: large-area printing of quantum dot is still at infancy state. Current lab scale printing using spin coating is not compatible with manufacturing. Electro-spraying deposition would be developed for high uniformity; 3) thin film encapsulation for QLED: current thin film encapsulation for flexible OLED requires multiple layers to ensure reliability during cycle bending. By evaluating the requirement of water vapor transmission rate of QLED and developing rather simplified material and structure, flexible QLEDs could be more cost effective.

3.3 Photomedicine demonstration with rigid and flexible QLED devices

The preliminary PBM and PDT results will pave the way for applying the bright, pure color red QLEDs in rigid or flexible form factors to positively change phototherapy applications in dermatology, oncology, minimally invasive surgery, stroke and brain disease among other fields.[1]

In the following section, detailed discussions on two critical photomedical fields (wound repair and cancer treatments) will be provided based on medical expertise of the authors' teams. With 660-nm QLEDs, laboratories involved in PBM therapy would be able to investigate wound healing, cellular metabolism and cell proliferation in vitro using multiple cell lines and in animal models of incisional and pressure ulcer wounds.[11] The effect of spectral response, dosimetry and power density on the rate and quality of healing could be evaluated by histology, collagen content and other parameters. The results would be incorporated in future institutional review board-approved human clinical trials to achieve healing of chronic wounds or acceleration of healing of primary wounds using surgical incisions such as hernia incisions, C-sections or breast augmentations.

It had been discovered by Drs. Hamblin and Huang that PF can be transformed into highly effective light-activated antimicrobials by adding a harmless solution of potassium iodide (KI). The 630-nm QLEDs could be used in vitro to eradicate antibiotic resistant strains of Gram-positive, Gram-negative bacteria and fungi using mixtures of PF and KI. Researchers could also test QLEDs in combination with PF and KI in a mouse model of an infected abrasion wound caused by the bioluminescent drug-resistant pathogen, *Acinenetobacter baumannii.*

Laser-based PDT has previously been successfully transferred into the clinic in the treatment of head and neck and lung cancer by the Shafirstein laboratory with the computer-simulation-assisted optimization of light delivery and dosimetry.[12–16] Similar methods can be applied to model light propagation from QLEDs emitting 630, 635 and 665 nm for PDT with PF, 5-ALA and 2-1 [hexyloxyethyl]-2-devinylpyropheophorbide-A, respectively. In Fig. 5, we present preliminary results from computer simulation of light propagation in a tissue mimicking geometry irradiated with 630-nm light wavelength delivered from a QLED with the model described previously in Oakley et al.[17] In this model, the light source was represented as a flux of photons emitted from the surface of the QLED. The resulting fluence rate (mW/cm^2) was computed throughout a cylindrical phantom, which mimicked the optical properties of tissue, when exposed to an input power density of 10 mW/cm^2. Figure 5 shows the models of the QLED and the phantom that were used and the resulting fluence rate (mW/cm^2) along two cross sections of the phantom (these cross sections are indicated A and B in Fig. 5b). The simulations suggest that the attenuation of the light emitted from a QLED will depend on the relative location within the device. Thus, the light power will attenuate to 60% of its maximum value in the middle of the phantom (line C, Fig. 5c) and 30% of its maximum value in the middle of the light emitted square (line D, Fig. 5d) at a depth of about 4 mm. In a laser-based PDT with a 8×8 mm source, the light will attenuate to 35% of its maximum value at the center of the laser beam at a depth of 4 mm in the same phantom (data not shown).

The evaluation of 635-nm QLEDs for ALA-PpIX PDT and associated dosimetry of oral malignancies can be carried out using a combination of in-vitro 3D co-culture models and animal models of oral cancers. This work will build on the prior experience of the Celli group in development and evaluation of LED-based PDT sources,[9] but leveraging QLEDs as a uniquely efficient and flexible source for ergonomic light delivery to lesions on otherwise challenging surfaces within the oral cavity.

Overall, as envisioned in these specific examples, the QLED light source could provide an ideal platform for various photomedicine applications.

4 Potential impact

The demonstrations of ultrabright deep red QLEDs and their effectiveness for PBM and PDT warrant further studies investigating QLED devices for photomedical applications. We believe that this QLED-based technology, initially developed out of strong interests from information display industry, can make inroads into new medical areas, enable the widespread clinical application and acceptance of photomedicine (e.g. PDT and PBM) and have a direct, beneficial impact to mankind by radically changing the ways to manage cancer, acute and chronic wounds, inflammation and antimicrobial resistance among others, with an estimated annual market size of over $200 billions.[18,19] The experience and knowledge gained in these developments could help enhance QLED stability and open up wider applications for QLEDs in display and lightings in the future.

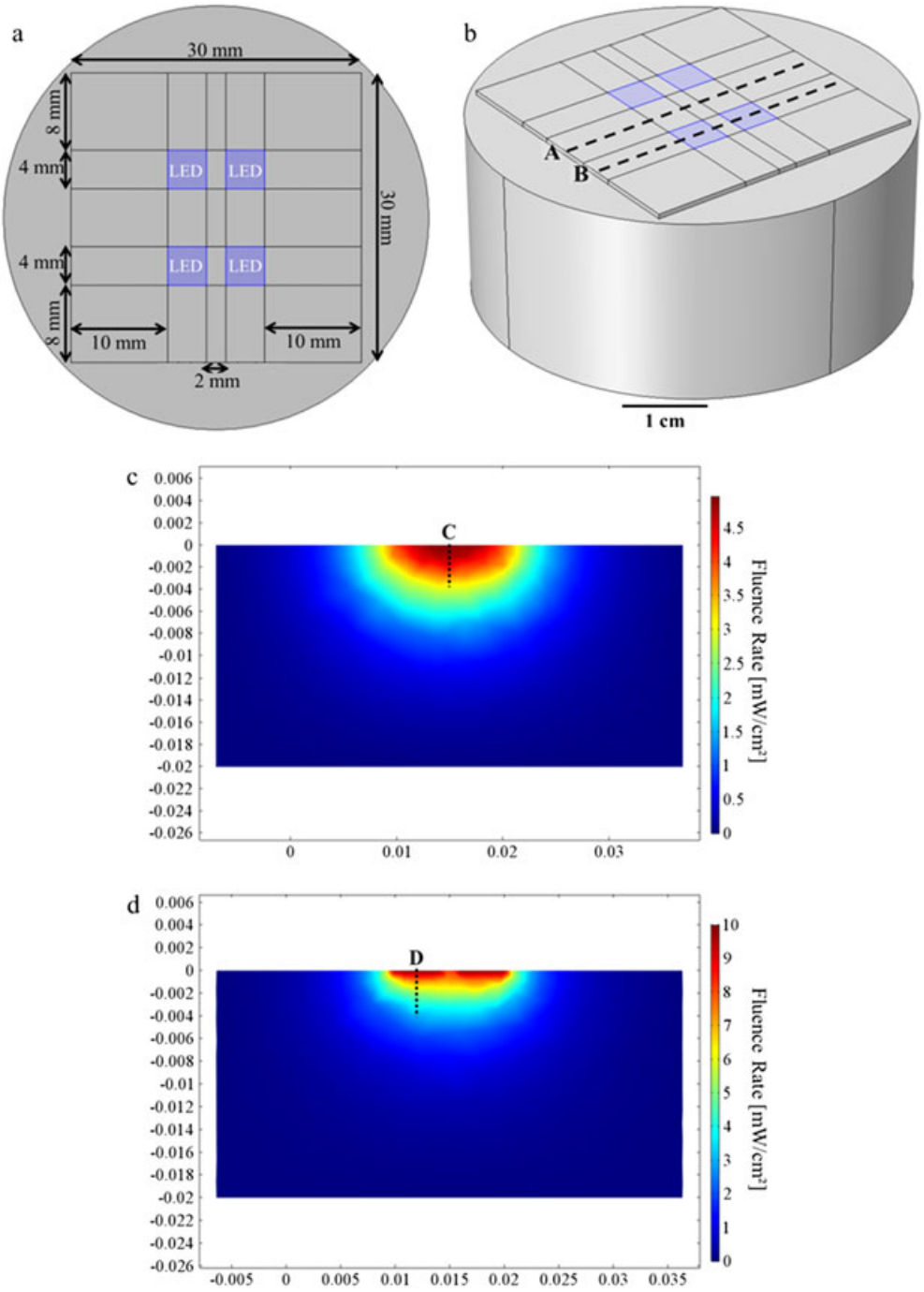

FIGURE 5 — Computer simulation of light propagation from a QLED in a tissue mimicking phantom. (a) Top view of the phantom and QLED. The purple squares indicate the location of the LEDs. The light source is modeled as a flux of photons emitted from these LEDs. (b) Geometry of tissue mimicking phantom with QLED placed on top. The phantom had dimensions of 22-mm radius and 20-mm height. (c) Resulting fluence rate (mW/cm^2) along one cross section of the phantom (line A from Fig. 5b). (d) Resulting fluence rate (mW/cm^2) along one cross section of the phantom (line B from Fig. 5b).

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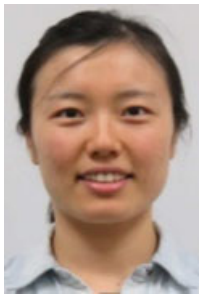
Hao Chen received his B.S. degree in Optical Information Science and Technology from Huazhong University of Science and Technology in 2011, his M.S. degree in Physical Electronics from Wuhan Research Institute of Posts and telecommunications in 2014 and is currently working toward the Ph.D. degree from the College of Optics and Photonics, University of Central Florida, Orlando. His current research in Dr. Yajie Dong's group focuses on QLEDs material and device development and their applications in display, lighting and photomedicine.



Juan He received her B.S. and M.S. degrees in the Department of Physics from Peking University, Beijing, China, in 2011 and 2014, respectively. She is currently working toward the Ph.D. degree at College of Optics and Photonics, University of Central Florida, Orlando, FL, USA. Her research focuses on quantum dots and other active materials for display, sensing and lasing application. She has published seven journal papers and three conference proceedings. She was a recipient of SID Distinguished Student Paper Award in 2016. She is the president of IEEE Photonics Society student chapter at University of Central Florida in 2016.

Raymond Lanzafame, M.D., M.B.A., F.A.C.S. received a Bachelor of Science with Honors and Distinction from Cornell, M.D. from George Washington University, and MBA from the Simon School of University of Rochester. He has specialized expertise in credentialing, medicolegal consulting, lasers and safety, and holds key leadership and Board positions in multiple professional and scientific organizations and task forces, including consultant to the General and Plastic Surgery Devices Advisory Committee of FDA. Publications include over 250 papers and three textbooks. His research interests include light–tissue interactions, imaging, host-tumor immunity, wound healing, photobiomodulation, photodynamic therapy, biomaterials, medical device and product development, technology assessment, clinical outcomes and education.

Istvan Stadler is a Research (Surgical) Scientist of Laser Center at Rochester General Hospital. He has a Ph.D. degree of Biochemistry/Biophysics from the School of Natural Sciences, Eotvos Lorand University, Budapest Hungary, and is Research Assistant Professor in Medicine at SUNY Buffalo School of Medicine. His research focuses on biological mechanisms of photobiomodulation, developing new treatment modalities for impaired wound healing and evaluating surgical and biomedical applications of laser technologies. His research is published in 70 peer-reviewed publications and in 72 presentations at national meetings. His translational research has received international attention of doctors, dentists, veterinarians, research scientists among others, providing new basic information on photobiomodulation mechanisms.

Hamid El Hamidi received his B.S. degree in Physics in 2005 from Hassan II University, Casablanca Morocco, and his M.S. degree in Applied Physics in 2014 from the University of Massachusetts Boston and currently working towards a Ph.D. degree in Applied Physics. He joined Prof. Celli's Photomedicine lab in 2012 where his research focuses on the influence of the mechanical microenvironment of solid tumors in photodynamic therapy response.

Hui Liu received her B.S. degree in Optoelectronics at Tianjin University. She pursued master and Ph.D. degrees in Applied Physics at Cornell University, where she studied nonlinear optics and ultrafast fiber lasers. She is experienced with designing and building various fiber lasers and calibrating the ultrafast pulses. She joined Professor Celli's group at the University of Massachusetts Boston from March 2015, working on a LED medical light source system development for photodynamic therapy in global health setting.

Jonathan Celli is an Assistant Professor in the Department of Physics at the University of Massachusetts Boston. His lab is focused on research at the intersection of cancer biology, biophysics and photomedicine, and is pursuing several projects on photodynamic therapy for cancer treatment. Prior to joining the University of Massachusetts, Prof Celli was junior faculty at the Wellman Center for Photomedicine at Massachusetts General Hospital and Harvard Medical School.

Michael R. Hamblin Ph.D. is a Principal Investigator at the Wellman Center for Photomedicine, Massachusetts General Hospital, an Associate Professor of Dermatology, Harvard Medical School and affiliated faculty at Harvard-MIT Division of Health Science and Technology. He directs a laboratory of around a dozen scientists who work in photodynamic therapy and photobiomodulation. He has published 346 peer-reviewed articles, is Editor or Associate Editor for 10 journals and serves on NIH Study-Sections. He has an h-factor of 77 and over 21 700 citations. He has authored/edited 11 proceedings volumes together with 10 other major textbooks on PDT and photomedicine. Dr Hamblin was honored by election as a Fellow of SPIE in 2011 and received the first Endre Mester Lifetime Achievement Award from NAALT in 2017.

Yingying Huang, M.D., Ph.D., is an instructor at Harvard Medical School in Dr. Hamblin's laboratory. Dr. Huang received her M.D. degree from Xiangya Medical School of Central South University. She received her M.S. degree in dermatology in Sun Yat-sen University. She received her Ph.D. degree in pathology from Guangxi Medical University. Dr. Huang's research interests lie in photodynamic therapy (PDT) for infections, cancer and mechanisms of low-level light therapy (LLLT) on injured cortical neurons. She has published over 60 peer-reviewed articles and 15 conference proceedings and book chapters.

Emily Oakley received a B.S. degree in Biomedical Engineering in 2013 from The George Washington University and a M.S. degree in Electrical Engineering in 2016 from the University at Buffalo. She is currently working on her Ph.D. degree in Molecular and Cellular Biophysics and Biochemistry at the Roswell Park Graduate Division of the University at Buffalo. Her research involves modeling the light propagation in tumors during interstitial PDT.

Gal Shafirstein is a Professor of Oncology, Department Cell Stress Biology, Roswell Park Cancer Institute (RPCI). He is the director of the photodynamic therapy (PDT) clinical research at RPCI. Dr. Shafirstein earned his Doctor of Science (D.Sc.) degree from the Technion, Israel Institute of Technology. His current research focuses on modeling light propagation in tissues and light dosimetry, and developing Phase I and II clinical studies using PDT in the treatment of patients with head and neck or lung cancer. Dr. Shafirstein holds six US patents and has published 69 papers in peer-reviewed journals. The National Cancer Institute of the National Institutes of Health, USA, funds his research.

Ho-Kyoon (H.K.) Chung is currently Chair Professor of Sungkyunkwan University. He was previously a full-time Advisor and Executive VP for Samsung SMD, which is dedicated to the R&D and manufacturing of AMOLED products and mobile LCD modules. Prior to that, he was the Executive Vice President and the head of Samsung SDI Corporate R&D Center from 2006 to 2008. Dr. Chung led the R&D for Active Matrix OLED (AMOLED) since 2000 and is a world leader in bringing the AMOLED technology into the commercial market. Prior experience includes Managing Director of Memory Product Engineering and Manufacturing Technology Center at Samsung Electronics, Semiconductor Business from 1988 to 1999.

Shin-Tson Wu is a Pegasus professor at College of Optics and Photonics, University of Central Florida. He is among the first six inductees of the Florida Inventors Hall of Fame (2014), and a Charter Fellow of the National Academy of Inventors (2012). He is a Fellow of the IEEE, OSA, SID and SPIE, and the recipient of 2014 OSA Esther Hoffman Beller medal, 2011 SID Slottow-Owaki prize, 2010 OSA Joseph Fraunhofer award, 2008 SPIE G. G. Stokes award and 2008 SID Jan Rajchman prize. Presently, he is serving as SID honors and awards committee chair.

Yajie Dong is an assistant professor in NanoScience Technology Center (NSTC) of University of Central Florida, with joint appointment in College of Optics and Photonics, and Department of Materials Science and Engineering. He received his B.S. and M.S. degrees in Chemistry from Tsinghua University of Beijing, China, and his Ph.D. degree from Harvard University. Before joining NSTC of UCF in August 2014, he worked as a Senior Scientist at QD Vision Inc and a postdoc associate at Massachusetts Institute of Technology. He is an associate editor of Optics Express and a member of Emissive Display (EMD) subcommittee of SID Technical Program Committee.