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Influence of Surface Energy and Roughness on Hole Mobility in Solution-Processed Hybrid Organic Thin Film Transistors

SUNGSIK LEE [1,2] (Member IEEE), FLORA M. LI [2,3], AND AROKIA NATHAN [2] (Fellow IEEE)

1 Department of Electronics Engineering, Pusan National University, Pusan 46241, South Korea
2 Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K.
3 Department of Information Technology, Apple Inc., Cupertino, CA 95014, USA

CORRESPONDING AUTHORS: S. LEE AND A. NATHAN (e-mail: sungsiklee@pusan.ac.kr; an299@cam.ac.uk)

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ABSTRACT We investigate the influence of the surface energy and roughness on the hole mobility in organic thin-film transistors where a poly(3,3'''-dialkylquarterthiophene) (PQT-12) hybridized with an octyltrichlorosilane self-assembled monolayer is employed as the semiconducting layer on a silicon nitride (SiN_x) gate insulator. Here, these surface properties are modified with varying the duration of oxygen plasma treatments on the SiN_x surface, eluding to a different surface roughness and energy. From our analysis coupled with the experimental results, it is found that the surface roughness (R_a) controls the degree of surface roughness scattering (χ_{SR}) while the surface energy (E_S) determines the reference mobility (μ_b), yielding the effective hole mobility expressed as a product of two terms associated with μ_b and χ_{SR}, respectively. It is found that μ_b follows a power law as a function of E_S, and χ_{SR} grows exponentially with increasing R_a. In addition, a characteristics scattering length (λ_c) appears in the mobility expression, which turns out to be a demarcation, suggesting that R_a is required to be at least smaller than λ_c to minimize χ_{SR}.

INDEX TERMS Organic thin film transistors, surface roughness scattering, surface energy, hole mobility.

I. INTRODUCTION

Organic semiconductors are one of the promising candidates for large area applications since they can meet a mechanical flexibility and low-temperature processability [1]–[5]. The latter mainly stems from the ability of a solution-based processing and ink-jet printing [6], [7]. In an organic device fabricated in such processes, an electrical performance, e.g., hole mobility, largely relies on the quality of the dielectric-semiconductor interface besides the degree of structural disorder in the semiconductor layer [8], [9]. Here, the quality of the dielectric-semiconductor interface and the molecular ordering of the organic semiconductor layer have a strong bearing on the field-effect hole mobility. Those issues are also severe in inorganic materials formed with a solution-based fabrication process. Here, the role of surface properties is crucial to determine an inorganic material-based transistor performance [10], [11]. As an example of organic materials, a poly(3,3'''-dialkylquarterthiophene) (PQT-12) TFT with a silicon nitride (SiN_x) gate insulator can have different interfacial properties and respective hole mobilities, depending on a dielectric surface treatment by either inserting a self-assembled monolayer (SAM) or applying an oxygen (O_2) plasma treatment on the SiN_x surface [12], [13]. Here, a quantitative investigation is required to physically explain the influences of these surface properties affect the hole mobility.

In this letter, we present a quantitative analysis on how surface roughness (R_a) and surface energy (E_S) physically affect the hole mobility in solution-processed organic TFTs. In the examined device, we employed a PQT-12 organic semiconductor layer hybridized with the octyltrichlorosilane (OTS) SAM on the SiN_x gate insulator. Here, as the key factor to vary the interfacial properties, the O_2 plasma treatment is performed on the SiN_x gate insulator surface, whose duration is experimentally varied to modify the SiN_x surface yielding different R_a and E_S. In our analysis coupled with the results from these experiments, we assume the distribution of the surface roughness follows a Gaussian probability function with the mean value of R_a while introducing a characteristics scattering length (λ_c). Besides, we use the Owens-Wendt method to retrieve E_S from the contact angle of water on the OTS SAM surface. Based on this, the modelled results indicate that R_a determines the degree of the surface roughness scattering (χ_{SR}) which grows exponentially with increasing R_a, and the reference mobility (μ_b) is governed by E_S following a power-law. These yield the hole mobility relation as $\mu_b(1 - \chi_{SR})$. In addition, it is suggested that λ_c can be treated as a demarcation to determine how much R_a needs to be reduced to minimize the surface roughness scattering.

II. RESULTS AND DISCUSSION

A. DEVICE PREPARATIONS

The examined TFTs were fabricated on a highly-doped silicon substrate which serves as the gate along with thermally-evaporated Cr-Au source/drain contacts and spin-coated PQT-12 as the semiconductor layer, as shown in Fig. 1(a). In particular, SiN_x as the gate dielectric was formed by plasma enhanced chemical vapour deposition (PECVD) at 150°C for a plastic substrate compatibility. After the deposition of SiN_x, the O_2 plasma treatment was carried out using a reactive ion etching (RIE) system with a chamber pressure of 150 mTorr and RIE power of 34 W. After that, the OTS SAM was formed by immersing the substrates in a 0.1M solution of OTS in toluene for 20 min at 60°C, followed by rinsing with toluene and isopropanol. These surface treatments were done prior to deposition of the PQT-12 layer. Electrical characterization of the OTFTs was carried out with a KEITHLEY 4200-SCS parameter analyzer. Surface properties were characterized using contact angle measurements for surface wettability and atomic force microscopy (AFM) measurements for surface roughness.

B. DIFFERENT SURFACE TREATMENTS AND HOLE MOBILITY

Fig. 1(b) shows the drain current (I_{DS}) vs. gate voltage (V_{GS}) measured at -60V drain voltage (V_{DS}) of PQT-12 OTFTs with 4 different types of surface treatments on SiN_x gate dielectric. Note that the amplitude of drain and gate voltages can be reduced with usage of a larger gate-insulator capacitance which can scale-down the threshold voltage, thus a low voltage operation [14], [15]. With this saturation I-V data, field-effect mobility (μ_{FE}) is retrieved with the following relation,

$$\mu_{FE} = \frac{1}{0.5(W/L)C_{ox}} \left(\frac{d\sqrt{|I_{DS}|}}{dV_{GS}} \right)^2, \tag{1}$$

where W is the channel width, L channel length, and C_{ox} gate-insulator capacitance. Also, an effective hole mobility (μ_{eff}) is captured as the peak value of μ_{FE} for each

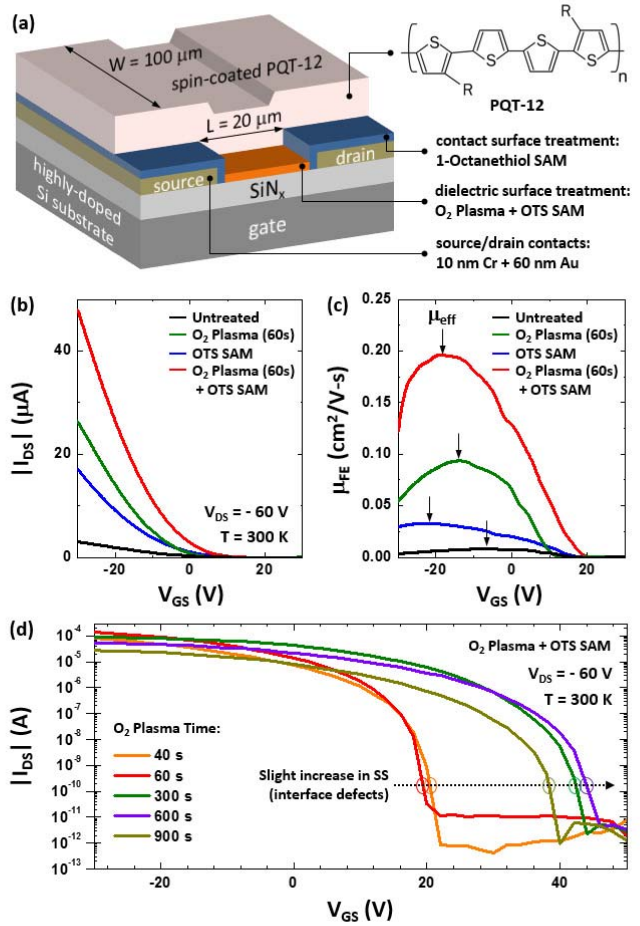

FIGURE 1. **(a) Schematic cross-section of the hybrid PQT-12 OTFT structure considered in this study. (b) Saturation transfer characteristics (i.e., I_{DS} vs. V_{GS}) measured at $V_{DS} = -60V$ for different surface treatments.**
(c) Field-effect mobility (μ_{FE}) retrieved from the measured I-V. Here, the effective hole mobility (μ_{eff}) is captured as a peak value for each case.
(d) I_{DS} vs. V_{GS} measured at $V_{DS} = -60V$ for the case of the O_2 plasma + OTS SAM while varying the oxygen plasma time.

case. As seen in Fig. 1(c), μ_{eff} is increased with the 60s O_2 plasma treatment. Furthermore, the O_2 plasma treatment coupled with the OTS SAM (i.e., O_2 plasma + OTS SAM) provides the largest μ_{eff}. This suggests that the combined treatment (O_2 plasma + OTS SAM) is one of the optimum ways to maximize the hole mobility further than the case for the O_2 plasma treatment only. For a further optimization of this case, the devices by the O_2 plasma + OTS SAM were further tested with varying the O_2 plasma time as seen in Fig. 1(d). Here, it is found that the sub-threshold slope (SS) is slightly increased with treatments for a longer plasma time > 60s, as retrieved in Fig. 2(a). This suggests that the interface quality is slightly degraded with a longer plasma time. For the above-threshold transfer characteristics, using Eq. (1), the μ_{eff} is extracted as a function of O_2 plasma duration (see Fig. 2(b)). A peak in μ_{eff} was observed at 60 s duration, which represents a 6.3-fold increase in mobility compared to devices without O_2 plasma exposure

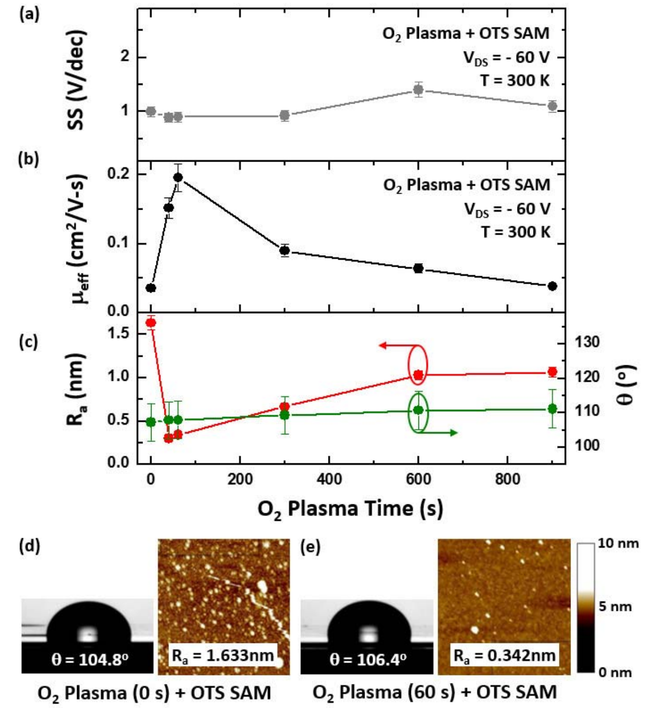

FIGURE 2. (a) Sub-threshold slope (SS) and (b) Effective hole mobility (μ_{eff}) as a function of the O_2 plasma time on SiN_x surface prior to the OTS SAM formation. Here, 5% error-bars are shown. (c) Water contact angle (θ) and mean value of the surface roughness (R_a) as a function of the O_2 plasma duration along with 5% error-bars. (d) θ and AFM images for the O_2 plasma exposure time = 0 s and (e) θ and AFM images for the O_2 plasma exposure time = 60 s, respectively.

(i.e., exposure time = 0 s). For longer exposures (t > 60 s), μ_{eff} gradually decreases. To understand these results in terms of surface properties, we examined the contact angle and surface roughness for the sample by the O_2 plasma + OTS SAM. Fig. 2(c) shows that the examined samples have a large contact angle (θ) > 100° which is slightly changed with a different oxygen plasma time (see also Figs. 2(d) and (e)). This is mainly attributed to a hydrophobicity induced with the O_2 plasma treatment [16]–[18], and a hydrophobic surface of the OTS SAM [19]. As can be seen, a more distinctive correlation is observed between surface roughness (R_a) and device mobility (μ_{eff}) in comparison with the contact angle. This implies that the surface energy is not so affected with the oxygen plasma. As illustrated in Figs. 2(b) and (c), there is a clear dependence of μ_{eff} on R_a. Here, we observe an inverse behavior between μ_{eff} and R_a as a function of the O_2 plasma time. At the plasma time ∼60 s, we have the maximum μ_{eff} and almost minimum R_a. Here, it is suggested that the oxygen plasma duration of 40∼60 s is an optimum. However, a longer plasma treatment gives rise to an over- etching while making R_a turn-around.

C. SURFACE ENERGY AND REFERENCE MOBILITY

Evident from the above discussion is a dependence of mobility on contact angle and surface roughness. Here, we develop a mathematical model to explain the relationships between these parameters. To account for contact angle effects in the mobility model, we first determine the surface energy (E_S) value from the contact angle (θ) data. To calculate E_s from θ, we used the Owens-Wendt method suitable for an OTS SAM surface [19], [20]

$$1 + \cos\theta = \frac{2}{E_L}\left[\left(E_S^d E_L^d\right)^{0.5} + \left(E_S^h E_L^h\right)^{0.5}\right], \quad (2)$$

where E_L^d is the liquid's component of dispersion force, E_L^h the liquid's component of hydrogen bonding force, $E_L(= E_L^d + E_L^h)$ the total surface tension/surface energy of the liquid, E_S^d the surface dispersion force component, E_S^h the hydrogen bonding force component of the surface, and $E_S(= E_S^d + E_S^h)$ the surface energy. Assuming $E_S^d \approx \alpha E_S^h$ and the ratio $\alpha = 5$, E_S^d and E_S^h are calculated solving Eq. (2). Note that the ratio (α) between E_S^d and E_S^h on the same material is found to be constant for a narrow range of the contact angles [20], and a = 5 is empirically deduced for the OTS SAM surface. Using the contact angle data in Fig. 2(c), E_S is retrieved, as shown in Fig. 3(a). Here, $E_L^d = 21.8$ mN/m, $E_L^h = 51.0$ mN/m, and $E_L = 72.8$ mN/m for water contact on the OTS SAM surface [19]. Based on E_S, the reference mobility (μ_b) at the surface is empirically represented as a power-law with the bulk band mobility (μ_{bulk}) of PQT-12,

$$\mu_b = \mu_{bulk}\left(\frac{E_S}{\zeta}\right)^{-\gamma}, \quad (3)$$

where ζ is a reference surface energy and γ the power-law exponent. Here, the reference mobility (μ_b) may be associated with an interface defect density which is increased with a longer plasma time (see Fig. 2(a)). Using Eq. (3), μ_b is drawn as a function of E_S for $\zeta = 8.9$ mN/m and $\gamma = 0.78$. And μ_{bulk} is to be 0.5 cm^2/V-s. As shown in Fig. 3(b), it is found that μ_b is decreased with increasing E_S.

D. SURFACE ROUGHNESS SCATTERING AND EFFECTIVE MOBILITY

In addition, we describe the effective mobility (μ_{eff}) while considering surface roughness and energy,

$$\mu_{eff} = \mu_b \int_{-\infty}^{\infty} D_{SR}(x)\gamma_s(x)dx. \quad (4)$$

Here, γ_s(x) is the scattering factor along the channel depth (x), represented as $\gamma_s(x)=\exp(-x/\lambda_c)$, λ_c is the characteristic scattering length, and $D_{SR}(x)$ is the distribution of surface roughness along the channel depth (x). Note that μ_b in Eq. (4) is employed as the pre-factor where the surface energy effect is considered through Eq. (3). For the distribution of the surface roughness, we employed a Gaussian random distribution represented as,

$$D_{SR}(x) = \frac{1}{\sqrt{2\pi}\sigma_{SR}} \exp\left[-\frac{(x - R_a)^2}{2\sigma_{SR}^2}\right], \quad (5)$$

where R_a is the mean value of the surface roughness and σ_{SR} is the variance of the surface roughness. Solving Eq. (4) with

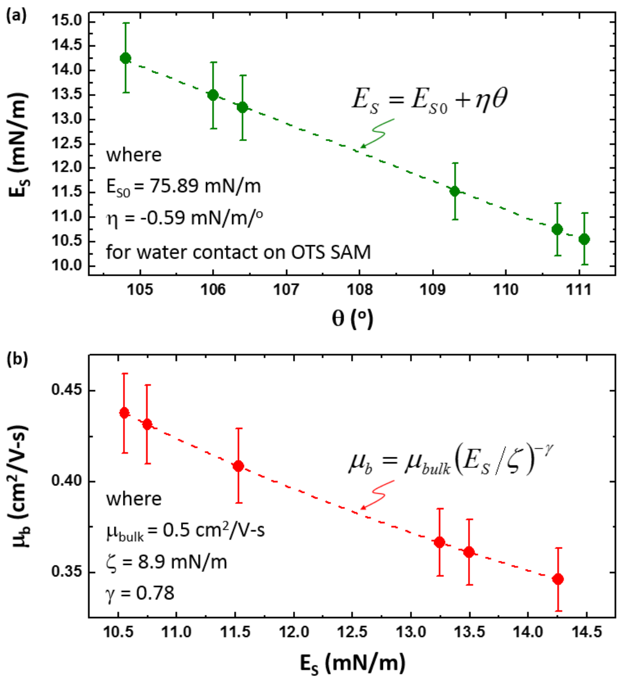

FIGURE 3. **(a) Surface energy (E_S) as a function of the water contact angle (θ), and (b) the reference mobility (μ_b) as a function of E_S on the OTS SAM surface, respectively. Here, E_{S0} is a constant as a intercept on the surface energy axis, and η is the slope with the contact angle. Here, 5% error-bars are shown.**

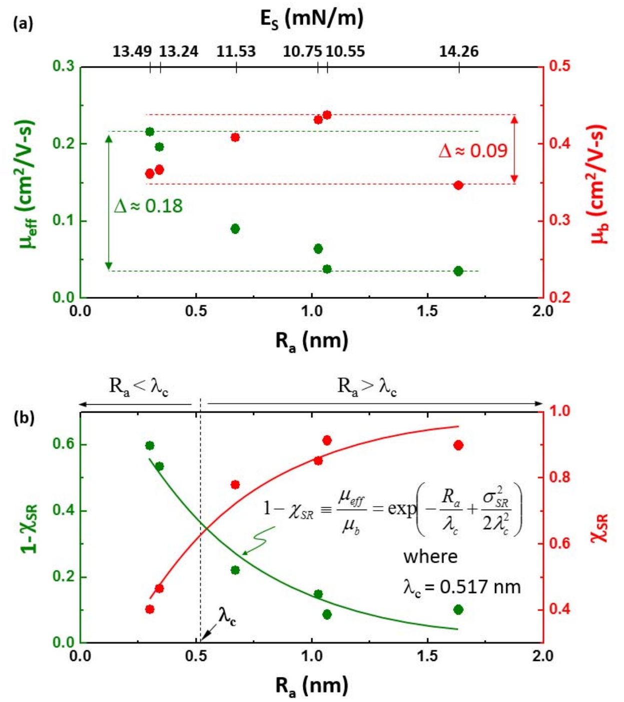

FIGURE 4. **(a) Effective hole mobility (μ_{eff}) and reference mobility (μ_b) as a function of the surface roughness (R_a). (b) 1-χ_{SR} (i.e., μ_{eff}/μ_b) and the degree of surface roughness scattering (χ_{SR}) as a function of R_a.**

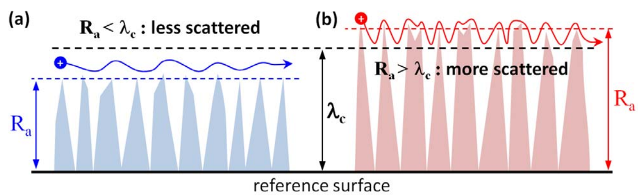

FIGURE 5. **Conceptual illustrations of the carrier movements when (a) the surface roughness (R_a) is smaller than the characteristic scattering length (λ_c) and (b) when R_a is larger than λ_c, respectively.**

Eq. (5), we can find an expression for the effective mobility (μ_{eff}) based on surface roughness (R_a) as well as surface energy (E_S),

$$\mu_{eff} = \mu_{bulk}\left(\frac{E_S}{\zeta}\right)^{-\gamma}\exp\left(-\frac{R_a}{\lambda_c}+\frac{\sigma_{SR}^2}{2\lambda_c^2}\right). \quad (6)$$

In order to capture the surface roughness effect, Eq. (6) can be rewritten as the ratio between μ_{eff} and μ_b,

$$\frac{\mu_{eff}}{\mu_b} = \exp\left(-\frac{R_a}{\lambda_c}+\frac{\sigma_{SR}^2}{2\lambda_c^2}\right) \equiv 1-\chi_{SR}, \quad (7)$$

where χ_{SR} is defined as the degree of surface roughness scattering. Note that the value of μ_{eff}/μ_b is increased if less scattered with the surface roughness, thus χ_{SR} is reduced. This implies an inverse proportionality between μ_{eff}/μ_b and χ_{SR}. Since the maximum value of μ_{eff}/μ_b is unity, it should be expressed as $1-\chi_{SR}$ rather than $1/\chi_{SR}$. Using the values of μ_{eff} and μ_b seen in Fig. 4(a), 1-χ_{SR} is plotted as a function of R_a, and a good agreement with the proposed model is achieved for $\lambda_c = 0.517$ nm while assuming $R_a >> \sigma_{SR}$, as shown in Fig. 4(b). Therefore, the mathematical model described here represents a practical means to quantitatively explain the dependence of mobility on both the surface energy and surface roughness.

Additionally, in the examined PQT-12 device treated with the O_2 plasma and OTS SAM, λ_c is found to be 0.517 nm, and χ_{SR} is less than 0.5 when $R_a < 0.517$ nm whereas it approaches unity when $R_a > 0.517$ nm, as seen in Fig. 4(b). This suggests that λ_c can be a demarcation for R_a. These discussions are conceptually illustrated in Fig. 5. Here, we believe that this kind of the demarcation with λ_c provides a quantitative guideline on how much R_a needs to be reduced for a higher mobility.

III. CONCLUSION

A quantitative analysis showed that the effective hole mobility in the examined PQT-12 thin film transistor hybridized with the OTS SAM and oxygen plasma treatments is composed of the product of the reference mobility and the surface roughness scattering-related terms. Here, the degree of the surface roughness scattering is inversely proportional to the surface roughness through their exponential relationship derived based on the Gaussian distribution along with the concept of the characteristics scattering length, and the reference mobility follows a power-law as a function of the surface energy. In particular, the introduced parameter, a characteristics scattering length, can be treated as a demarcation on how much the surface roughness needs to be reduced for a higher mobility. And it was suggested that the surface roughness is required to be scaled to be at least smaller than the characteristics scattering length to reduce the surface roughness scattering, yielding a higher mobility. In the transistor examined here, the characteristics scattering length is found to be 0.517 nm for the OTS SAM PQT-12. Consequently, the analysis presented here gives quantitative and analytical insights into surface properties, which determine the electrical performance and stability of thin film transistors.

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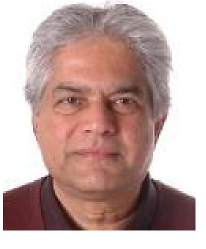
SUNGSIK LEE received the Ph.D. degree from the University College London, London, U.K., in 2013. He has been a Professor with Pusan National University, Pusan, South Korea, since 2017. From 2013 to 2017, he was a Research Associate with the University of Cambridge, Cambridge, U.K.. His area of expertise is semiconductor devices and physics for futuristic electronics. He has published over 70 articles and with an *H*-index of 17. He was a recipient of the IEEE EDS Ph.D. student fellowship 2011 and the Best Teaching Prize 2017 from the Korean Society for Engineering Education, South Korea.

FLORA M. LI received the Ph.D. degree in electrical engineering from the University of Waterloo, specialized in thin film electronics based on metal oxide semiconductors, organic semiconductors, and nanomaterials. She is currently a Multidisciplinary Researcher and a Program Manager with Apple, passionate about developing new technologies and solutions to make the world a better place. Her specialties lie in research, fabrication process development, material science, engineering, device/material characterization, circuit design and testing, system integration, and project management.



AROKIA NATHAN holds the Chair of photonic systems and displays with the Department of Engineering, Cambridge University. He was a Post-Doctoral Fellow with LSI Logic Corporation, USA, and ETH Zurich, Switzerland, for several years. He joined the University of Waterloo, ON, Canada, where he held the DALSA/NSERC Industrial Research Chair in sensor technology and, subsequently, the Canada Research Chair in nano-scale flexible circuits. In 2006, he became the Sumitomo Chair of nanotechnology with the London Centre for Nanotechnology, University College London. He has held Visiting Professor appointments with the Physical Electronics Laboratory, ETH Zurich, and the Engineering Department, Cambridge University, U.K. He has published over 500 papers in the field of sensor technology, CAD, and thin-film transistor electronics, and has co-authored four books. He has over 70 patents filed/awarded and has founded/co-founded four spin-off companies. He was a recipient of the Royal Society Wolfson Research Merit Award from University College London. He serves on technical committees and editorial boards in various capacities. He is a Chartered Engineer, U.K., a fellow of the Institution of Engineering and Technology, U.K., and an IEEE/EDS Distinguished Lecturer.