Theory of signal and noise in double-gated nanoscale electronic pH sensors

Jonghyun Go, Pradeep R. Nair,^{a)} and Muhammad A. Alam

School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907, USA

(Received 8 April 2012; accepted 20 June 2012; published online 13 August 2012)

The maximum sensitivity of classical nanowire (NW)-based pH sensors is defined by the Nernst limit of 59 mV/pH. For typical noise levels in ultra-small *single-gated* nanowire sensors, the signal-to-noise ratio is often not sufficient to resolve pH changes necessary for a broad range of applications. Recently, a new class of *double-gated* devices was demonstrated to offer apparent “super-Nernstian” response (>59 mV/pH) by amplifying the original pH signal through innovative biasing schemes. However, the pH-sensitivity of these nanoscale devices as a function of biasing configurations, number of electrodes, and signal-to-noise ratio (SNR) remains poorly understood. Even the basic question such as “Do double-gated sensors actually resolve smaller changes in pH compared to conventional single-gated sensors in the presence of various sources of noise?” remains unanswered. In this article, we provide a comprehensive numerical and analytical theory of signal and noise of double-gated pH sensors to conclude that, while the theoretical lower limit of pH-resolution does not improve for double-gated sensors, this new class of sensors does improve the (instrument-limited) pH resolution. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4737604>]

I. INTRODUCTION

Field effect devices for biomolecule detection and pH sensing have been explored since the 1970's¹ for potential applications in healthcare, food safety, and environmental monitoring. With the reduction in device dimensions, nanoscale FET devices such as nanowire (NW) or nanoplate (NP) sensors² promise ultrasensitive detection, ease of integration, and lower-cost fabrication and, therefore, continue to attract significant research interest. In the constant current mode, the pH-sensitivity (S) of FET-based sensor is defined as the change in gate voltage (ΔV_G) necessary to restore the current following a change in pH (ΔpH), i.e., $S = (\Delta V_G)/(\Delta pH)$. In principle, single-gated pH-sensors can achieve the maximum pH sensitivity defined by the Nernstian response of 59 mV/pH; however, in practice electrolyte screening and semiconductor capacitance³ reduces the sensitivity considerably below the theoretical limit. Moreover, relatively high noise floor of ultra-scaled NW sensors⁴ limits the signal-to-noise ratio (SNR) and the corresponding pH resolution ($\Delta pH_{\min}$) of such devices. Here, we define $\Delta pH_{\min}$ as being the minimum change in pH above the noise floor that can be continuously (without signal averaging) detected by the FET-sensor.

It is therefore not surprising that there has been considerable interest in amplifying the signal for higher pH sensitivity. Indeed, one may view Beckman's original pH meters, which amplified the signal from electro-chemical pH probes by a vacuum diodes, as the first effort in this direction.⁵ The ease of use of these pH meters revolutionized the field; however, since both signal and noise are amplified simultaneously, the larger signal of a Beckman pH-meter did not translate to better

pH resolution. More recently, a number of groups^{6–9} have reported experimental observation of “super-Nernstian” amplification in a variety of systems, most notably in double-gated field-effect transistors or DGFETs.¹⁰ However, the theory of pH sensitivity of these sensors as a function of device biasing configuration and device geometry remains poorly understood. As a result, there is no simple way to compare the performance of various device geometries that report Super-Nernstian response and evaluate whether one scheme is intrinsically superior to others. Most importantly, the fundamental question of whether these amplification schemes simultaneously improve SNR and reduce pH resolution remains unresolved. More generally, this ambiguity in the theory of pH-sensitivity of DGFETs is also reflected in our inability to predict if the sensitivity and SNR will continue to improve with the number of gate electrodes. In other words, if two electrodes of a DGFET improve sensitivity, will a tri-gated or a quadruple gated device perform even better?

In this article, we offer a comprehensive theoretical analysis of double-gated pH sensors, with emphasis on the “so-called” amplified Nernstian response and the signal-to-noise ratio. We combine the classical theory of the MOSFET (Ref. 11) and the site-binding model¹² to calculate the signal amplification and the noise levels for DGFETs in various biasing configurations (i.e., accumulation, depletion, and inversion). The theoretical work provides a unifying framework to interpret a broad range of experimental data reported in the literature.^{6–9} *Rather surprisingly and somewhat counter-intuitively, we find that if both signal and noise can be measured with infinite precision, the best pH-resolution achieved by a DGFET sensor is indistinguishable from that of a single-gated sensor.* On the other hand, if the minimum detectable pH change is determined by the noise associated with inexpensive instrumentation—as sometimes is the case

^{a)}Present address: Department of Electrical Engineering, Indian Institute of Technology Bombay, Mumbai, India.

for commercial systems—DGFETs will offer better signal to noise ratio and improved pH -resolution compared to classical counterparts.

The article is arranged as follows. Section II describes the model system, then we validate the model in Sec. III by comparing with experimental results for single-gated ion-sensitive field effect transistors (ISFETs). An analytical theory of double gate pH sensors with emphasis on super-Nernstian response and the noise analysis are described in Secs. IV and V, respectively. These results are subsequently supported by numerical simulation and available experimental data from the literature. Finally, we discuss the implications of the results and summarize our conclusions in Sec. VI.

II. MODEL SYSTEM

A. System description

A simplified diagram of the single, double, and multiple gate field-effect transistors is shown in Fig. 1. The system generally consists of a nanoscale FET, whose traditional metal/polysilicon gate is replaced by an electrolyte and a reference electrode immersed in it.¹ This reference electrode (typically Ag/AgCl) establishes a fixed electrostatic potential (Ψ) inside the solution¹³ and allows application of a gate bias (V_{FG}) to the transistor.¹⁴ For a single-gated ISFET, the Si-body is grounded, and the drain (D) is biased slightly positive with respect to the source (S). Any change in pH is reflected in the S-to-D channel current. For a DGFET, there

is an additional gate at the bottom of the transistor (V_{BG}), which is coupled to the channel through a thicker bottom oxide. Finally, in a FinFET-like multi-gated configuration (MGFET), the additional gates may be independently controlled by various biasing configurations (V_{BG1} , V_{BG2} , etc.) These additional gates may allow flexible settings of optimum bias condition. We will focus operation of ISFETs and DGFETs but will also have a few comments regarding MGFET.

B. Model equations

For the following analysis, the DGFET can be divided into four regions (see Fig. 1(d) and the associated equations are provided in Table I): (i) fluid gate-electrolyte interface described by Eq. (1), (ii) the electrolyte described by Eq. (2), (iii) the top oxide-electrolyte interface described by Eq. (3), and finally, (iv) the oxide-Si FET system described by Eqs. (4)–(8). The corresponding equations for noise are summarized in Table II, through Eqs. (9)–(11). And, the symbols and the numerical values used in this paper are summarized in Table III. We now describe the physical motivation of using these specific equations in some detail.

1. Fluid gate-electrolyte interface

An important assumption made in the traditional model (see Eq. (1)) is that the reference electrode is *faradaic*: A faradaic electrode can exchange electrons with ions in electrolyte such that there is no potential drop at the electrode-electrolyte interface, and the electrostatic potential applied at the electrode drops fully at the other side of the electrolyte (i.e., the sensor surface in case of ISFET sensors), as illustrated in Eq. (1) of Table I.¹³ A non-faradaic electrode, on the other hand, is characterized by an insulating interface between electrolyte and electrode surface so that it involves charging interface (electric double layer) with no transfer of electrons across the interface. Thus, the potential applied at the non-faradaic electrode drops fully near the electrolyte-electrode interface and the electrode cannot control the electrostatics at the sensor surface. We will restrict ourselves to systems that obey Eq. (1) and will not discuss the anomalous pH -response associated with partially-faradic gate.^{15,16} We assume that the noise associated with the faradaic fluid-gate is negligible,⁴ so that there is no corresponding entry for noise in Table II. However, this may not be case for partially-faradic electrodes,¹⁷ and will need careful consideration.

2. Electrolyte system

The ions in bulk electrolyte are assumed to be 1:1 (NaCl or KCl) and the distribution of cations (n_+) and anions (n_-) follows the Boltzmann distribution, see Eq. (2). Note that the fluid-gate bias is coupled to Eq. (2) through the exponent. The Poisson equation is used to calculate the overall potential within the electrolyte system. The noise associated with thermal fluctuation within the electrolyte is summarized in Eq. (9), Table II. The electrolyte bulk resistance (R_b) in Eq. (9) is given by Ref. 4, based on the assumption that the

FIG. 1. Schematics of (a) a conventional ISFET sensor, (b) a double-gate FET (DGFET) sensor, and (c) a multi-gate FET (MGFET) sensor. The device dimensions and physical parameters can be found in Table III. The four different regions of the sensor discussed in Sec. II is illustrated in (d). The model equations of each region are summarized in Tables I and II.

electrolyte is NaCl. This electrolyte noise level, however, is generally much smaller than the low-frequency noise of MOSFETs (which will be discussed later).

3. Top oxide-electrolyte interface

At the electrolyte-top gate oxide interface, the surface of the top oxide can be functionalized with surface groups ($-\text{OH}$), which protonate and deprotonate as a result of the reactions with protons (H^+) in electrolyte so that the net charge of OH groups respond to the change of pH of the solution. We assume that the protonation/deprotonation of OH groups are dictated by the surface binding model, see Eq. (3),^{12,18} treated within the Poisson-Boltzmann approximation and use the well-known values for surface group density and the protonation/de-protonation constants from the literature (see Table III). The model is based on continuum approximation and effects associated with finite-size of the molecules are neglected. We anticipate future refinement of the model based on more sophisticated approaches.

4. Signal and noise in FETs

The electrostatics of Si body is governed by Poisson-Boltzmann equation, see Eq. (5). Equations (4)–(6) are sufficient to describe classical ISFET. For a DGFET sensor, Eqs.

(7) and (8) must also be solved self-consistently to define the potential within the silicon body. In this model, we have assumed that the sensing device has a planar geometry so that we can solve 1-D Poisson-Boltzmann equations, and for simplicity, the differences of work function between different materials consisting of the ISFET/DGFET sensor are not considered explicitly. In calculating *ideal* sensitivity of ISFET/DGFET sensors, we assume that the density of interface traps at the gate oxide-Si interface¹¹ (governed by Eq. (5) in Table I) is negligible, but we do consider their presence to explain the experimental data to be discussed in Sec. III. Indeed, as expected, the conclusions were found to be not sensitive to such details of the model.

In addition to the electrolyte noise discussed above, a classical ISFET pH sensor has several different noise sources such as thermal noise and $1/f$ noise. Given the typical measurement duration of pH sensors, we only need to consider the low frequency noise ($1/f$) from MOSFETs. To predict the noise and SNR we follow the number-mobility fluctuation model.⁴ The corresponding noise of ISFET is given by Eq. (10). For the noise calculation in DGFET sensors, we account for the noise sources from the top and bottom sides of Si body by assuming that the DGFET can be viewed as two independent MOSFETs with different oxide thicknesses and gate voltages. Therefore, the noise of the combined

TABLE I. The model equations at each region (illustrated in Fig. 1(d)) of ISFET/DGFET sensors.

Devices	Region	Equations
ISFET/DGFET region (i)	Fluid gate-electrolyte interface	$\Psi _{\text{FG-elec}} = V_{\text{FG}}$ (V_{FG} : fluid gate bias) (1)
ISFET/DGFET region (ii)	Electrolyte	$-\nabla \cdot (\epsilon_w \nabla \Psi) = q(n_+ - n_-)$ $n_+ = n_0 \exp(-q(\Psi - V_{\text{FG}})/k_B T)$ $n_- = n_0 \exp(q(\Psi - V_{\text{FG}})/k_B T)$ (2)
ISFET/DGFET region (iii)	Top oxide-electrolyte interface	$(\epsilon_{\text{ox}} \nabla \Psi _{\text{tox-elec},0^-}) - (\epsilon_w \nabla \Psi _{\text{tox-elec},0^+}) = \sigma_{\text{OH}}$ $\sigma_{\text{OH}} = qN_s \frac{10^{pK_a - \text{pH}} e^{-\beta \psi_0} - 10^{\text{pH} - pK_b} e^{\beta \psi_0}}{1 + 10^{pK_a - \text{pH}} e^{-\beta \psi_0} + 10^{\text{pH} - pK_b} e^{\beta \psi_0}}$ (3) <p>where $\psi_0 \equiv \Psi _{\text{tox-elec}} - V_{\text{FG}}$ (Ref. 18).</p>
ISFET/DGFET region (iv)	Top gate oxide	$-\nabla \cdot (\epsilon_{\text{ox}} \nabla \Psi) = 0$ (4)
ISFET/DGFET region (iv)	Interface sites	$Q_{it} = qD_{it}(E_g/2 - k_B T/q \times \log(N_A/n_i) - \Psi)$ (5)
ISFET/DGFET region (iv)	Si Body	$-\nabla \cdot (\epsilon_{\text{Si}} \nabla \Psi) = q(p - n - N_A)$ (6)
DGFET region (iv)	Bottom gate oxide	$-\nabla \cdot (\epsilon_{\text{ox}} \nabla \Psi) = 0$ (7)
DGFET region (iv)	bottom oxide-gate interface	$\Psi _{\text{BG-box}} = V_{\text{BG}}$ (V_{BG} : bottom gate bias) (8)

TABLE II. The model equations of noise analysis in ISFET/DGFET sensors.

Electrolyte noise	
$S_{V_e} = 4kTR_b$	
$\delta V_e = \sqrt{\int_{f_1}^{f_2} S_{V_e} df} = \sqrt{4kTR_b(f_2 - f_1)}$	(9)
Gate voltage noise in ISFET sensors	
$\delta V^{ISFET}(V_{FG}) = \sqrt{\int_{f_1}^{f_2} S_{V_{FG}} df}$	(10)
$S_{V_{FG}(BG)} = S_{V_{FB,top}(bot)} \left[1 + \left(\alpha \mu_{eff} C_{eff,top(bot)} \frac{I_{DS,top(bot)}}{g_{m,top(bot)}} \right)^2 \right]$	
$S_{V_{FB,top(bot)}} = \frac{q^2 k T N_t \lambda}{f W L C_{eff,top(bot)}^2}, g_{m,top(bot)} = \frac{dI_{DS,top(bot)}}{dV_{FG}(BG)}$	
Gate voltage noise in DGFET sensors	
$\delta V_{BG}^{DGFET}(V_{FG}) = \sqrt{\int_{f_1}^{f_2} S_{I_{DS}}^{DGFET} / g_{m,bot}^2 df _{V_{BG}}}$	(11)
$= \sqrt{\int_{f_1}^{f_2} \left(\frac{g_{m,top}^2}{g_{m,bot}^2} S_{V_{G,top}} + S_{V_{G,bot}} \right) df _{V_{BG}}}$	
$S_{I_{DS}}^{DGFET}(V_{FG}) \equiv S_{I_{DS,top}} + S_{I_{DS,bot}} _{V_{BG}}$	
$= g_{m,top}^2 S_{V_{FG}} + g_{m,bot}^2 S_{V_{BG}} _{V_{BG}}$	

device can be determined as an uncorrelated sum of the noises from top and bottom channels. This assumption will have to be revised for ultrathin body, fully depleted DGFET transistors. The overall noise voltage seen from the bottom gate is given by Eq. (11). Note that for the DGFET, the pH sensitivity depends on the bias regime of fluid gate (the detailed analysis will be provided in Sec. IV); likewise, the drain current noise and gate voltage noise in Eq. (11) depend on the fluid gate bias (V_{FG}). We use the compact model of conventional MOSFET (Ref. 11) to calculate the effective capacitance ($C_{eff,top}$ and $C_{eff,bot}$), the channel current ($I_{DS,top}$ and $I_{DS,bot}$), and the transconductance ($g_{m,top}$ and $g_{m,bot}$). In previous studies, the effective capacitance (C_{eff}) was replaced by the gate oxide capacitance,⁴ but here we define C_{eff} as $C_{eff} = d(-Q_s)/dV_G$, where Q_s is the total charge per unit area induced in the silicon. This redefinition explicitly accounts for the depletion capacitance, so that one may predict the complete bias-dependence (i.e., depletion and inversion) of the ISFET/DGFET sensor noise. We emphasize that there is no new contribution to the development of theoretical models in Tables I and II: we simply use these well-established models from the literature within a self-consistent theoretical framework to explore the effects of device geometry and bias conditions on pH sensitivity and noise of ISFET/DGFET sensors.

The equations in Table I are first solved self-consistently to obtain electrostatic potential (Ψ) of every region in the ISFET/DGFET system. The corresponding carrier concentration (n) is obtained based on the electro-

static potential profile (Ψ) inside the Si body. The current (I_{DS}) flowing through the Si body is computed with the expression $I_{DS} = \mu n W V_{DS}/L$ —where μ , W , and L is the mobility, width, and length of the channel, respectively—assuming ISFET/DGFET sensors are long channel devices with a small source-drain bias (V_{DS}). Once the DC operating condition related to a given biasing condition, pH , and salt concentration is determined, the equations in Table II allow us to calculate the noise associated with the operating conditions.

III. MODEL VALIDATION BY INTERPRETING RESPONSE OF A SINGLE-GATED NW pH SENSORS

We begin by interpreting the response of a nanowire pH sensor (Fig. 2), as a function of fluid-gate bias. The pH sensitivity of traditional ISFETs, biased at various fluid gate voltages, can be understood simply as follows (see Ref. 3 for detailed analysis): The amphoteric OH groups at gate oxide/buffer undergo protonation/deprotonation of interface as a function of surface proton density, $[H^+]_s$. Assuming Boltzmann distribution for ions in buffer, we have $[H^+]_s = [H^+]_B e^{-q\psi_0/k_B T} = e^{-2.303 pH - q\psi_0/k_B T}$, where $[H^+]_B$ is the bulk proton density, $pH = -\log_{10}[H^+]_B$, and ψ_0 is the potential difference developed at the oxide-buffer interface. As the surface proton density $[H^+]_s$ dictates the surface charge on the oxide, we approximately assume that $[H^+]_s$ remains constant under the constant-current operation of the ISFET. The corresponding potential change due to the pH shift (ΔpH) is thus given by $\Delta\psi_0 \approx 2.303(k_B T/q) \Delta pH$. Hence, the

TABLE III. The definition of symbols and their default values used in the equations summarized in Tables I and II.

Definition	Symbol	Default value
Sensor width	W	100 nm
Sensor length	L	10 μm
Top gate oxide thickness	t_{tox}	4 nm
Bottom gate oxide thickness	t_{box}	150 nm
Si body thickness	t_{Si}	80 nm
Si doping density	N_A	10^{15} cm^{-3}
Si intrinsic carrier density	n_i	$1.1 \times 10^{10} \text{ cm}^{-3}$
Si carrier mobility	μ_{eff}	$100 \text{ cm}^2/\text{V s}$
Drain voltage	V_{DS}	0.1 V
Vacuum permittivity	ϵ_0	$8.85 \times 10^{-14} \text{ F/cm}^2$
Silicon permittivity	ϵ_{Si}	$11.8 \epsilon_0$
Oxide permittivity	ϵ_{ox}	$3.9 \epsilon_0$
Water permittivity	ϵ_w	$80 \epsilon_0$
Interface trap density	D_{it}	$10^9 \text{ cm}^{-2} \text{ eV}^{-1}$
Si bandgap	E_g	1.1 eV
Surface group density	N_s	$5 \times 10^{14} \text{ cm}^{-2}$
Protonation constant	pK_a	-2
De-protonation constant	pK_b	6
Electrolyte strength	n_0	100 mM
Electrolyte resistance	R_b	$3.31 \times 10^4 \Omega$
Volumetric trap density	N_t	$3 \times 10^{16} \text{ eV}^{-1} \text{ cm}^{-3}$
Tunneling parameter	λ	0.5 Å
Coulomb scattering coefficient	α	$1.5 \times 10^5 \text{ V s/C}$
Frequency band	f_1, f_2	0.01 Hz, 1000 Hz

maximum pH sensitivity, known as the Nernst limit, is $\Delta\psi_0/\Delta pH = 59 \text{ mV}/pH$. Due to the absence of potential drop inside the bulk electrolyte (as we assume a faradaic fluid gate), the corresponding shift in the fluid gate bias (V_{FG}) is also given by $\Delta V_{FG}/\Delta pH \simeq 59 \text{ mV}/pH$. In reality, the sensitivity is always less than the Nernst limit (e.g., $40 \text{ mV}/pH$ in Fig. 2), due to the high electrolyte screening,^{19,20} protonation affinity of sensor surface, and finite semiconductor capacitance,³ as reflected in the following numerical interpretation of the experimental data.

In a fluid-gated FET sensor, changes of electrolyte pH (Fig. 1(a)) and/or sweeping fluid gate bias (V_{FG}) modulate the conductance of Si body. Solving Eqs. (1)–(5) yields the corresponding current-bias (I_{DS} - V_{FG}) characteristics with different pH values, as shown in Fig. 2. The variation in pH results in modulation of the net charge at the top oxide-electrolyte interface via protonation/deprotonation of surface OH groups and is reflected as a potential shift in the transfer characteristics. We also calibrated our model and demonstrated the consistency of simulation results with the recent experiment data of Si nanowire (SiNW) FET pH sensors. The pH sensitivity is defined as the shift of threshold voltage ($V_{T,FG}$) due to pH changes ($\Delta V_{T,FG}/\Delta pH$) in constant-current operation. Note that the pH sensitivity is always less than the well-known Nernst limit ($59 \text{ mV}/pH$), although the specific values depend on various parameters like the surface properties, physical dimensions, and operating conditions. In particular, the measured pH sensitivity from experiments, $40 \text{ mV}/pH$, is consistent with our theoretical calculation (inset of Fig. 2). Further, note that our model reproduces experimental results in both the depletion and the inversion regimes using a single set of parameters—indicating that the model is predictive. We now extend this model to address the response of DGFET sensors (Sec. IV) followed by the noise comparison between ISFET and DGFET sensors in Sec. V.

IV. AMPLIFIED NERNST SIGNAL AND SNR OF DOUBLE GATE FETS

A. Key features of experimental observations

As discussed in the introduction, many recent publications suggest that the limit of $59 \text{ mV}/pH$ can be breached and an amplified Nernst sensitivity $\sim 1 \text{ V}/pH$ can be achieved by using the double-gated, asymmetric silicon-on-insulator structures (DGFETs, Fig. 1(b)). In DGFET sensors, one sweeps the bottom gate (BG) bias with the fluid gate (FG) bias held fixed, to obtain the transfer characteristics (I_{DS} - V_{BG}). The corresponding pH sensitivity is measured in terms of the threshold voltage shift ($\Delta V_{T,BG}/\Delta pH$). The conductance change at the top surface of channel due to pH shift ($\Delta I_{t,FG}$) is compensated by conductance change at the bottom surface ($\Delta I_{b,BG}$) to maintain constant current operation ($\Delta I_{t,FG} + \Delta I_{b,BG} = 0$).

The origin of high pH sensitivity in DGFET sensors has been attributed to the asymmetry between top and bottom oxide thicknesses, and the amplification factor has been related to the capacitance ratio of the top and bottom gate oxides, such that $\Delta V_{BG}/\Delta pH = \alpha_{SN}(\Delta V_{FG}/\Delta pH) \times (C_{tox}/C_{box})$.

FIG. 2. The transfer characteristics of a single-gate FET pH sensor (with grounded bottom contact) with different values of pH from experiments from our collaborators (red and black circles) as well as the numerical simulations results (red and black solid lines) from the calibrated theoretical model. The inset shows the corresponding V_T shifts ($\Delta V_{T,FG}$) with respect to pH from experiments (circles) and simulations (solid line). The slope of the blue lines is the pH sensitivity ($\sim 40 \text{ mV}/pH$ in this particular case). The calibrated model uses the following physical parameters: $N_A = 10^{16} \text{ cm}^{-3}$, Si thin body thickness is $t_{si} = 70 \text{ nm}$, the top gate oxide thicknesses are $t_{tox}(\text{SiO}_2) = 4 \text{ nm}$ and $t_{tox}(\text{Al}_2\text{O}_3) = 9 \text{ nm}$, bottom gate oxide thickness $t_{box} = 150 \text{ nm}$, and salt concentration is $n_0 = 50 \text{ mM}$. We also use the well-known values for pK_a , pK_b , and OH group density (N_S) for Al_2O_3 -electrolyte interface: $pK_a = 6$, $pK_b = 10$, $N_S = 8 \times 10^{14} \text{ cm}^{-2}$ and the interface trap density is assumed to be $D_{it} = 4.1 \times 10^{11} \text{ cm}^{-2} \text{ eV}^{-1}$. The experimental data are given in Ref. 22.

Remarkably, however, while some group find $\alpha_{SN} \simeq 1$,^{6,7} others report $\alpha_{SN} \ll 1$ (Refs. 8 and 9) (which may entirely negate the improvement due to capacitance ratio!). This discrepancy appears to be related to the fluid gate bias, but a comprehensive theory of this difference has not been offered. In the following section, we provide a systematic numerical analysis of the bottom gate and fluid gate sensitivities of a DGFET pH sensor and explain how the response depends on C_{tox}/C_{box} ratio corresponding to the top/bottom oxide scaling as well as the biasing regimes of the DGFET. We validate the numerical results with simple analytical formulation and show that the experimental results from different research groups can be interpreted systematically within this theoretical framework.

B. Numerical model of DGFET pH -sensors

Two types of experiments have been reported in the literature: pH sensitivity as a function of thickness of the bottom oxide (t_{box}) for a fixed fluid-gate bias (see Figs. 3 and 5(a)) and pH -sensitivity as a function of fluid-gate bias (V_{FG}) for fixed device geometry (Figs. 4 and 5(b)). To predict the pH response of DGFET sensors for fixed FG-bias (e.g., $V_{FG} = 0 \text{ V}$), we first solve the equations in Table I self-consistently and obtain the I_{DS} - V_{BG} characteristics for different pH values. We repeat the numerical calculations with several different bottom oxide thicknesses to check the dependency of ΔV_{BG} on t_{box} . The simulation results (Fig. 3) are consistent with the recent experimental studies⁹ in which the fluid gate biases the channel to depletion regime, and the

scaling of ΔV_{BG} shows linearity with respect to t_{box} , as previously reported in the literature.⁸

Similarly, to explore the pH response of a device for a given geometry with variable FG-bias (second type of experiments reported in the literature⁹), we solve the equations in Table I self consistently for different fluid gate biases (with parameters summarized in Table III), while keeping the BG-bias large enough so that the bottom channel is in inversion. The result is an S-shaped response, plotted in Fig. 4. The pH sensitivity for fixed device geometry (i.e., fixed C_{tox}/C_{box}) depends on the FG bias conditions: while at low FG-bias (i.e., depletion regime in minority carrier devices), the response is small and relatively flat (<0.3 V), it goes through a transition at the intermediate FG-bias (~ 0.3 - 0.8 V) and eventually levels off at high FG bias (>0.8 V, corresponding to inversion regime). Figs. 3 and 4, taken together, explain the experimental observation that the signal amplification in DGFETs depends on the product of capacitance ratio (C_{tox}/C_{box}) and a bias-dependent (V_{FG}) factor. *In the following discussion, however, we show that this widely held perception is strictly true for scaling of bottom-gate oxide* (Figs. 5(a) and 5(b)); *a more nuanced analysis is necessary for top-gate scaling* (Fig. 5(c)).

C. Analytical theory of DGFET sensors

To understand the physical origin of linear response on oxide thickness in Fig. 3 and the bias-dependent response in Fig. 4, let us consider a simple analytical argument. For DGFET sensors, a pH induced potential change at top oxide/buffer interface (ΔV_{FG}) modulates the inversion charge of DGFETs by $\Delta I_{t,FG} \sim C_T \Delta V_{FG}$, where C_T denotes the capacitive coupling between the buffer and Si channel. Under constant current operation, the conductance change at top channel needs to be compensated by an equivalent modulation of bottom channel conductance. Assuming same mobility for top and bottom channels, this leads to $-\Delta I_{t,FG} = \Delta I_{b,BG} \sim C_B \Delta V_{BG}$, where C_B denotes the capacitive coupling between the bottom gate and Si channel. Accordingly, pH sensitivity during bottom gate operation is

$$\frac{\Delta V_{BG}}{\Delta pH} = \frac{\Delta V_{FG}}{\Delta pH} \times \left(\frac{C_T}{C_B} \right). \quad (12)$$

Equation (12) applies for all bias regimes of DGFETs provided C_T and C_B are properly defined. Here we consider the two different regimes of practical DGFETs operation (the sensitivity of all possible bias regimes are shown in Sec. I of the Supplementary Information):²¹

- (i) *Regime I (inversion-inversion regime)*, in which both the top and bottom channels of the DGFET are in inversion. In this regime (i.e., with high V_{FG}), $C_T = C_{tox}$, $C_B = C_{box}$.
- (ii) *Regime II (depletion-inversion regime)* in which the top channel is under depletion (i.e., with low V_{FG}) and bottom channel is under inversion. Here, $C_B = C_{box}$ while C_T involves additional contribution due to the Si body capacitance. Accordingly, we have C_T

FIG. 3. The threshold voltage shift ($\Delta V_{T,BG}$) with respect to pH changes (ΔpH) in a bottom gate operation with three different bottom oxide thicknesses (t_{box}). The slopes of green, blue, and red curves are 85, 165, and 240 mV/pH, respectively, and the red circles indicate the experimental observation in the literature.⁹ The inset shows the correlation of pH sensitivity ($\Delta V_{T,BG}/\Delta pH$) with t_{box} which shows linear scaling relationship. Here, we assume an undoped Si body, t_{tox} is 4 nm and t_{box} is 50, 100, and 150 nm, respectively, and t_{Si} is 80 nm. We also use the well-known values for pK_a , pK_b , and OH group density (N_S) for Al_2O_3 -electrolyte interface: $pK_a = 6$, $pK_b = 10$, $N_S = 8 \times 10^{14} \text{ cm}^{-2}$.

$= C_{tox} C_{Si} / (C_{Si} + C_{tox})$, where C_{Si} is the thin body Si capacitance ($C_{Si} = \epsilon_{Si} / t_{Si}$).

Since ΔV_{FG} follows the Nernst response ($\Delta V_{FG}/\Delta pH \leq 59 \text{ mV/pH}$), the DGFET pH sensitivity is given as

$$\frac{\Delta V_{BG}}{\Delta pH} = \frac{\Delta V_{FG}}{\Delta pH} \times \left(\frac{C_{tox}}{C_{box}} \right) \times \alpha_{SN}, \quad (13a)$$

where α_{SN} ($0 < \alpha_{SN} \leq 1$) is a factor depending on the bias conditions at the top and bottom surface:

FIG. 4. The pH sensitivity ($\Delta V_{T,BG}/\Delta pH$) in dual gate operation ($V_{FG} \neq 0$ V) as a function of V_{FG} . We assume a DGFET sensor with the same physical parameters as the ones shown in Fig. 3 except t_{box} of 150 nm. The dotted lines indicate the pH sensitivity corresponding to the simple analytic expressions in Eq. (13b).

FIG. 5. Validation of geometry (t_{box} scaling) and bias dependence of sensitivity and its comparison with experimental data of DGFET sensors in the literature are shown in (a) and (b), respectively. The red lines are obtained from numerical calculations with appropriate parameters corresponding to each device demonstrated in Refs. 8 and 9. The blue lines are obtained by multiplying red lines with corresponding γ factors coming from Eq. (13b). (c) The scaling relationship of bottom gate sensitivity ($\Delta V_{\text{BG}}/\Delta \text{pH}$) with respect to t_{tox} scaling. Depending on the ratio ($C_{\text{tox}}/C_{\text{Si}}$) in Eq. (13b), the sensitivity in depletion-inversion regime (blue curve) shows different relationship with the top oxide scaling.

$$\alpha_{\text{SN}} = \begin{cases} 1 & (\text{regime I: inversion - inversion}) \\ (1 + C_{\text{tox}}/C_{\text{Si}})^{-1} & (\text{regime II: depletion - inversion}). \end{cases} \quad (13b)$$

Equation (13b) correctly anticipates the asymptotic limits of the curve shown in Fig. 4 (dashed horizontal lines). The reduced sensitivity in regime II reflects the fact that the depletion capacitance of the Si body, connected in series with C_{tox} , lowers the total capacitance C_{T} . Applying a high bias at the fluid gate (moving from regime II to regime I) creates the charge inversion layer at the top surface of the Si body, which electrostatically separates the top and bottom channels of the Si body and removes the body capacitance that degrades amplification. This dependency on V_{FG} indicates the amplification factor in pH sensitivity is not exclusively determined by $C_{\text{tox}}/C_{\text{box}}$ (in contrast to what has been claimed previously in the literature) but also is very sensitive to the bias conditions.

D. Quantitative interpretation of experimental data reported in the literature

Here, we interpret the experimental data from Refs. 8 and 9 using our analytical/numerical model. We first numerically calculate (by solving Eqs. (1)–(8) in Table I, see Figs. 5(a) and 5(b)) the bottom gate sensitivity of a DGFET sensor at high (red line) and low V_{FG} (blue line) regimes, as a function of thickness of the bottom gate oxide (t_{box}). Since the body-thickness (t_{Si}) and top-oxide thicknesses (t_{tox}) are held fixed, Eq. (13b) suggests that the responses should scale linearly with t_{box} and the low- V_{FG} response (i.e., at the depletion-inversion regime) should be suppressed by a constant factor, $\gamma \equiv \alpha_{\text{SN,regime II}}/\alpha_{\text{SN,regime I}} = (1 + C_{\text{tox}}/C_{\text{Si}})^{-1}$, with respect to the high- V_{FG} operation. These predictions are consistent with both features of the experimental data reported in Ref. 8, i.e., the linearity of response with bottom oxide thickness and the suppression factor $\gamma = (1 + C_{\text{SAM}}/C_{\text{ZnO}})^{-1} = (1 + (\kappa_{\text{SAM}}\epsilon_0/\kappa_{\text{ZnO}}\epsilon_0)/(t_{\text{SAM}}/t_{\text{ZnO}}))^{-1} = 0.254$, as the device with ZnO channel and self-assembled monolayer (SAM) as gate dielectric ($t_{\text{SAM}} \sim 2$ nm) was operated in the low V_{FG} regime. The suppression factor ($\sim 1/4$) was previously attributed to non-ideal process issues;⁸ however, our analysis reveals that it arises as a natural consequence of the low bias (V_{FG}) condition.

Likewise, the experiment from Ref. 9 can be easily interpreted based on the geometrical parameters of the device and the operating condition, as shown in Fig. 5(b). The sensitivity reaches the idealized maximum when biased in the inversion-inversion (high bias) regime, consistent with the theory discussed above. The suppression factor γ corresponding to the specific devices shown in Ref. 9 is equal to $(1 + C_{\text{tox}}/C_{\text{Si}})^{-1} \approx 0.764$, where $C_{\text{tox}} = C_{\text{SiO}_2}^{-1} + C_{\text{Al}_2\text{O}_3}^{-1} \approx 5.37 \times 10^{-8}$ F/cm², based on the dimensions of SiO₂-Al₂O₃ gate insulator. The measurement data (symbols) obtained in different bias conditions (V_{FG}) are consistent with the model prediction.

The aforementioned comparisons, however, do not highlight an interesting difference regarding the scaling of top vs. bottom gate oxides. In the *high-bias* (V_{FG}) regime, the back-gate sensitivity is exclusively given by $\Delta V_{\text{BG}}/\Delta \text{pH} \approx (\Delta V_{\text{FG}}/\Delta \text{pH}) \times (C_{\text{tox}}/C_{\text{box}})$, therefore scaling *up* of bottom oxide thickness is interchangeable with scaling *down* of

top-oxide. In the depletion-inversion regime (low bias), however, $\alpha_{SN} \approx (C_{tox}/C_{Si})^{-1}$ if $C_{tox} \gg C_{Si}$ (i.e., if $t_{Si} \gg t_{tox}$, see Eq. (13b)), so that $C_{tox} \times \alpha_{SN}$ is approximately constant, and therefore $\Delta V_{BG}/\Delta pH \approx (\Delta V_{FG}/\Delta pH) \times (C_{tox}/C_{box}) \times \alpha_{SN}$ remains invariant under t_{tox} scaling. In other words, in this regime, the sensitivity will no longer scale with capacitance ratio, as shown in Fig. 5(c). The regime has never been reported in the literature because experiments have been confined to bottom-gate scaling, with fixed top-gate and Si body thicknesses. Note that the linear scaling with capacitance ratio could be restored with thick top oxides ($t_{tox} \gg t_{Si}$) with $\alpha_{SN} \approx 1$.

In summary, our interpretation of the available experiments suggests that the DGFET sensors should be operated in inversion-inversion regime for maximum signal amplification. As we will discuss in the Sec. V, however, that regime of maximum signal amplification also coincides with high noise and correspondingly results in little enhancement in signal-to-noise ratio, making the design of DGFET pH-sensor a difficult optimization problem.

E. Potential of multi-gate pH-sensors

The analysis of DGFET sensors discussed above and its high pH sensitivity raise an interesting question: If a “single” bottom gate improves pH sensitivity, would pH sensors with multiple independent bottom gates (MGFET, See Fig. 1(c)) improve sensitivity even further? To answer this question, a detailed 2D modeling of MGFET’s electrostatics as well as its noise estimation is required. A detailed simulation using a 2D commercial device simulator (MEDICI) shows that there is no advantage of using additional bottom gates (see Sec. III of Supplementary Information for details) and the amplification offered by DGFETs is actually close to the maximum that can be achieved by multi-gate pH sensors. In other words, the pH amplification obtained by a MGFET sensor can always be replicated by a DGFET with suitably tailored device geometry and operating condition. Therefore, one may wish to adopt MGFET configuration for process compatibility with devices such as FINFET, but not for any intrinsic advantage of pH sensitivity.

V. SIGNAL TO NOISE RATIO OF DGFET SENSORS

The ultimate goal of amplifying pH sensitivity is to enhance the corresponding pH resolution, that is, to reduce the minimum pH change that can be detected by a sensor. The pH resolution in an electronic pH sensing scheme is defined as $\Delta pH_{\min} \equiv 3 \times \delta V_G/S$ (pH unit), where δV_G is the gate voltage noise (V unit), and S is the pH sensitivity ($\Delta V/\Delta pH$, V/pH unit); according the definition of $\Delta pH_{\min}$, devices offering lower $\Delta pH_{\min}$ can resolve smaller changes in pH. To improve $\Delta pH_{\min}$, one must not only improve the pH sensitivity, but also ensure that the noise is not amplified in proportion. In this section, we discuss the pH resolution of ISFET and DGFET sensors based the noise model we described in Sec. II (and in Table II).

Fig. 6 shows the results of noise calculation for ISFET and DGFET pH sensors. The current noise component

$S_{I_{ps}}^{DGFET}$, calculated from Eq. (11), are shown in Fig. 6(a). Fig. 6(c) compares the gate voltage noise of ISFET and DGFET pH sensors depending on the bias conditions. Similar to our analysis of bias dependence of pH-sensitivity as summarized in Eq. (13a), we again consider two different bias regimes (depletion vs. inversion regime in terms of V_{FG}) for the noise ISFET/DGFET sensors:

A. Noise in the low-bias, depletion-inversion regime

In the depletion regime of the ISFET, the term $\alpha \mu_{eff,top} C_{eff,top} I_{DS,top}/g_{m,top}$ in Eq. (10) is considerably smaller than 1, so that $S_{V_{G,top}} \simeq S_{V_{FB,top}}$ and $\delta V_{ISFET}^{ISFET} \sim 1/C_{eff,top} = 1/C_{tox} + 1/C_D$, where C_D is the Si depletion capacitance. For a DGFET sensor, biased in the depletion-inversion regime, $\delta I_{DS,top}^2 \ll \delta I_{DS,bot}^2|_{V_{BG}}$ (See Fig. 6(a)), thus Eq. (11)

$$\text{yields } \delta V_{BG}^{DGFET} \simeq \sqrt{\int_{f_1}^{f_2} S_{V_{G,bot}} df} \sim 1/C_{eff,bot}.$$

Since we fix the bottom gate bias in its inversion regime (the red dot in Fig. 6(a)), $C_{eff,bot} \simeq C_{box} (= \epsilon_{ox}/t_{box})$, therefore, $\delta V_{BG}^{DGFET} \sim 1/C_{box}$. Although $C_{tox} \gg C_{box}$ for typical DGFET dimensions, however, depending on the fluid gate bias (which modulates C_D), the noise of ISFETs $\delta V_{ISFET}^{ISFET} (\sim 1/C_{eff,top} = 1/C_{tox} + 1/C_D)$ can be larger than that of DGFETs $\delta V_{BG}^{DGFET} (\sim 1/C_{box})$ in a certain bias range (Fig. 6(b)). However, in low bias regime, they are of comparable magnitude, as shown in Fig. 6(b).

B. Noise in the high-bias, inversion-inversion regime

In the inversion regime for ISFET, $\alpha \mu_{eff,top} C_{eff,top} I_{DS,top}/g_{m,top} \gg 1$, so that δV_{ISFET}^{ISFET} is independent of $C_{eff,top} (= C_{tox})$. For a DGFET sensor in the inversion-inversion regime, from Eq. (11) $S_{V_{G,top}}$ is smaller than $S_{V_{G,bot}}$ but $(g_{m,top}^2/g_{m,bot}^2) S_{V_{G,top}} \gg S_{V_{G,bot}}$ because $g_{m,top}^2/g_{m,bot}^2 \sim (C_{tox}/C_{box})^2 \gg 1$. Therefore, $\delta V_{BG}^{DGFET} \sim C_{tox}/C_{box} \gg 1$, as a result, the DGFET noise is larger than that of ISFET by a factor of C_{tox}/C_{box} (i.e., $\delta V_{BG}^{DGFET} \approx (C_{tox}/C_{bot}) \times \delta V_{ISFET}^{ISFET}$, implying that the noise also scales with capacitance ratio!), as shown at the high bias regime in Fig. 6(b).

Finally, the pH resolution ($\Delta pH_{\min} \equiv 3 \times \delta V_G/S$) of ISFETs and DGFETs are obtained by replacing S with $\Delta V_{FG}/\Delta pH$ and $\Delta V_{BG}/\Delta pH$, respectively, and δV_G with δV_{ISFET}^{ISFET} and δV_{BG}^{DGFET} , respectively. Note that $\Delta V_{FG}/\Delta pH$ remains constant with respect to V_{FG} , while $\Delta V_{BG}/\Delta pH$ changes with V_{FG} (See Fig. 4(a)). The corresponding results are shown in Fig. 6(c). In depletion regime, the pH resolution of DGFETs is smaller than that of ISFETs due to two reasons as described in Sec. V A, (i) the noise levels are comparable for DGFETs and ISFETs (low bias regime of Fig. 6(b)), and (ii) the signal of DGFETs is larger by a factor of $(C_{tox}/C_{box}) \times \alpha_{SN} > 1$ (see Eq. (13a)) where $\alpha_{SN} = (1 + C_{tox}/C_{Si})^{-1}$. On the other hand, for the inversion regime, we have $\Delta V_{BG}/\Delta pH = \Delta V_{FG}/\Delta pH \times (C_{tox}/C_{box})$ and $\delta V_{BG}^{DGFET} \approx (C_{tox}/C_{box}) \times \delta V_{ISFET}^{ISFET}$. Note that as both the signal and noise of DGFETs scales with C_{tox}/C_{box} , the SNR is invariant under the bottom oxide scaling in inversion-

FIG. 6. (a) DGFET noise components of Eqs. (11) and (12) in Table II. The terms $\delta I_{DS,top}$, $\delta I_{DS,bot}|_{V_{BG}}$, and δI_{DS}^{DGFET} are given by $\delta I_{DS,top} = \sqrt{\int_{f_1}^{f_2} S_{I_{DS,top}} df}$, $\delta I_{DS,bot}|_{V_{BG}} = \sqrt{\int_{f_1}^{f_2} S_{I_{DS,bot}|_{V_{BG}}} df}$, and $\delta I_{DS}^{DGFET} = \sqrt{\int_{f_1}^{f_2} S_{I_{DS}}^{DGFET} df}$, respectively. (b) and (c) The corresponding noise and ΔpH_{min} of the ISFET and DGFET pH sensor.

inversion regime. Similarly the SNR of DGFETs in depletion-inversion regime does not scale with bottom oxide thickness, since the noise δV_{BG}^{DGFET} is also proportional to the bottom gate oxide thickness ($\delta V_{BG}^{DGFET} \sim 1/C_{eff,bot} = 1/C_{box} \sim t_{box}$).

To summarize the discussion above, we find that the DGFET pH sensors are superior to ISFET sensors in terms of SNR in their depletion regime (set by V_{FG}), and one can, in principle, achieve higher SNR and correspondingly resolve smaller changes in pH at depletion regime. In contrast, the performance of DGFET in the inversion-inversion regime is indistinguishable from that of an ISFET and remains invariant under the oxide scaling. Therefore, we conclude that the high signal amplification in the inversion-inversion regime may not lead to improved pH resolution.

C. Role of instrumentation noise

The noise analysis in Sec. V is based on the assumption that the noise of the measuring instrument (by which we can resolve the back-gate voltage signal) is infinitesimally small, and thus the pH measurement is not limited by the precision of the instrument but by the intrinsic noise from the DGFET transistor. In practice, it may be too costly to integrate high-precision measurement systems in low-cost, point-of-care pH sensors. If the extrinsic noise from measurement instrument (δV^{Ins}) is the dominant component, the SNR of a DGFET sensor operated at its inversion-inversion regime can be superior to that of an ISFET, as illustrated in Figure S1 and Table S1 of the Supplementary Information. The analysis shows that if the sensor's SNR is limited by the capability of the measurement instrument to discriminate voltage with given precision,

the noise of ISFETs/DGFETs becomes both comparable to δV^{Ins} , although the sensitivity of DGFETs still remains much higher than that of ISFET. This implies that, unless sophisticated and precise measurement instrument is available for signal detection, the DGFET can offer superior SNR and pH resolution regardless of its operational regime (see Table S1 in the Supplementary Information for details).

VI. CONCLUSIONS

In this paper, we have developed a numerical and analytical theory of amplified pH response of a DGFET sensor and have used the theory to consistently interpret a broad range of experimental observations. Based on the discussion above, we arrive at the following conclusions:

- (i) The maximum pH sensitivity for DGFET sensors can be achieved in the inversion-inversion regime and is given by the ratio of top and bottom oxide capacitance (C_{tox}/C_{box}).
- (ii) Theoretically, we show that the SNR of DGFET is higher (and thus gives lower pH resolution) compared to ISFET, if the fluid gate is biased in its depletion regime. Unfortunately, the pH resolutions become comparable when both are operated in the inversion regime.
- (iii) In practice, DGFETs may be viewed as a low-noise differential amplifier for the pH-signal. Therefore, if the noise from the measurement equipment sets the lower limit of sensor performance, a DGFET will always offer a better SNR compared to a single-gated ISFET.
- (iv) And finally, a close analysis of the electrostatics in multi-gate pH-sensors suggests that further

improvement in sensitivity or SNR is unlikely with additional independent gates.

ACKNOWLEDGMENTS

The authors acknowledge computational and financial support from Network of Computational Nanotechnology (NCN) and National Institute of Health (NIH). The authors also appreciate Professor T. Mayer in Penn state University for sharing experimental data and useful discussion regarding double-gated FET-based pH sensors.

- ¹P. Bergveld, "Thirty years of ISFETOLOGY: What happened in the past 30 years and what may happen in the next 30 years," *Sens. Actuators B* **88**(1), 1–20 (2003).
- ²Y. Cui, Q. Wei, H. Park, and C. M. Lieber, "Nanowire nanosensors for highly sensitive and selective detection of biological and chemical species," *Science* **293**(5533), 1289–1292 (2001).
- ³L. Bousse, N. F. De Rooij, and P. Bergveld, "Operation of chemically sensitive field-effect sensors as a function of the insulator-electrolyte interface," *IEEE Trans. Electron Devices* **30**(10), 1263–1270 (1983).
- ⁴M. J. Deen, M. W. Shinwari, J. C. Ranuárez, and D. Landheer, "Noise considerations in field-effect biosensors," *J. Appl. Phys.* **100**, 074703 (2006).
- ⁵A. O. Beckman and H. E. Fracker, "Apparatus for testing acidity," U.S. Patent 2,058,761 (1936).
- ⁶B. Khamaisi, O. Vaknin, O. Shaya, and N. Ashkenasy, "Electrical performance of silicon-on-insulator field-effect transistors with multiple top-gate organic layers in electrolyte solution," *ACS Nano* **4**(8), 4601–4608 (2010).
- ⁷M. Spijkma, J. J. Brondijk, T. C. T. Geuns, E. C. P. Smits, T. Cramer, F. Zerbetto, P. Stoliar, F. Biscarini, P. W. M. Blom, and D. M. de Leeuw, "Dual-gate organic field-effect transistors as potentiometric sensors in aqueous solution," *Adv. Funct. Mater.* **20**(6), 898–905 (2010).
- ⁸M. Spijkman, E. C. P. Smits, J. F. M. Cillessen, F. Biscarini, P. W. M. Blom, and D. M. de Leeuw, "Beyond the Nernst-limit with dual-gate ZnO ion-sensitive field-effect transistors," *Appl. Phys. Lett.* **98**, 043502 (2011).
- ⁹O. Knopfmacher, A. Tarasov, W. Fu, M. Wipf, B. Niesen, M. Calame, and C. Schönenberger, "Nernst limit in dual-gated Si-nanowire FET sensors," *Nano Lett.* **10**(6), 2268–2274 (2010).
- ¹⁰J. Go, P. R. Nair, B. Reddy, Jr., B. Dorvel, R. Bashir, and M. A. Alam, "Beating the Nernst limit of 59 mV/pH with double-gated nano-scale field-effect transistors and its applications to ultra-sensitive DNA biosensors," *Tech. Dig. – Int. Electron Devices Meet* **2010**, pp. 1–4.
- ¹¹Y. Taur and T. H. Ning, *Fundamentals of Modern VLSI Devices*, 2nd ed. (Cambridge University Press, 2009).
- ¹²S. L. David, E. Yates, and T. W. Healy, "Site-binding model of the electrical double layer at the oxide/water interface," *J. Chem. Soc., Faraday Trans. 70*, 1807–1818 (1974).
- ¹³J. Bard and L. R. Faulkner, *Electrochemical Methods: Fundamentals and Applications*, 2nd ed. (Wiley, New York, 2000).
- ¹⁴E. D. Minot, A. M. Janssens, I. Heller, H. A. Heering, C. Dekker, and S. G. Lemay, "Carbon nanotube biosensors: The critical role of the reference electrode," *Appl. Phys. Lett.* **91**(9), 093507 (2007).
- ¹⁵G. Tarabella, C. Santato, S. Y. Yang, S. Iannotta, G. G. Malliaras, and F. Cicoira, "Effect of the gate electrode on the response of organic electrochemical transistors," *Appl. Phys. Lett.* **97**, 123304 (2010).
- ¹⁶P. Lin, F. Yan, and H. L. W. Chan, "Ion-sensitive properties of organic electrochemical transistors," *ACS Appl. Mater. Interfaces* **2**(6), 1637–1641 (2010).
- ¹⁷A. Hassibi, R. Navid, R. W. Dutton, and T. H. Lee, "Comprehensive study of noise processes in electrode electrolyte interfaces," *J. Appl. Phys.* **96**(2), 1074–1082 (2004).
- ¹⁸D. Landheer, G. Aers, W. R. McKinnon, M. J. Deen, and J. C. Ranuarez, "Model for the field effect from layers of biological macromolecules on the gates of metal-oxide-semiconductor transistors," *J. Appl. Phys.* **98**(4), 044701 (2005).
- ¹⁹P. R. Nair and M. A. Alam, "Screening-limited response of nanobiosensors," *Nano Lett.* **8**(5), 1281–1285 (2008).
- ²⁰P. R. Nair and M. A. Alam, "Design considerations of silicon nanowire biosensors," *IEEE Trans. Electron Devices* **54**(12), 3400–3408 (2007).
- ²¹See supplementary material at <http://dx.doi.org/10.1063/1.4737604> for the discussions of the analytic derivation of bias-dependent pH sensitivity, the relationship between pH resolution and the instrumental noise, and the analysis of multi-gate FET (MGFET) sensors can be found in the supplementary material.
- ²²T. Mayer, personal communication (Penn State University, 2011).