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Reply to the 'Comment on "Electron-interfered field-effect transistors as a sensing platform for detecting a delicate surface chemical reaction"' by M. Micjan and M. Weis, *J. Mater. Chem. C*, 2026, 14, DOI: 10.1039/D5TC02689J

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Michal Micjan and Martin Weis argue that electron-interfered FET (EIFET) cannot detect surface reactions via threshold-voltage shifts (ΔV_{th}) because charge produced at the interference electrode (IE) must manifest primarily as a gate current (I_G); they further suggest that the reported V_{th} dynamics arise from bias-stress and interfacial trapping rather than chemistry at the IE. We rebut these claims on three grounds. First, a capacitive-network analysis shows that the transient displacement current, I_G , is quantitatively and operationally negligible, while the quasi-static V_{th} modulation is the only relevant observable. Charge transiently accumulated at the IE modulates the effective gate bias through capacitive coupling and produces a measurable, reversible $V_{th}(t)$ signal. Second, the characteristic $V_{th}(t)$ signature co-varies with an independent surface-science observable—the IE surface energy $\gamma_s(t)$ —and survives stringent controls (dielectric substitution, solvent/ionic solutions, and preformed SAMs) that a trap-only hypothesis cannot explain. Third, the magnitude and time scale of ΔV_{th} are quantitatively consistent with small, transient IE charge densities (10^{11} – 10^{12} cm⁻²), far below monolayer coverages and fully compatible with the proposed mechanism.

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1. Scope of the rebuttal and summary of the comment

The Comment advances three assertions: (i) electrons released during Au–thiol chemistry at the IE necessarily induce an equal and opposite image charge on the gate, implying that the primary observable should be a I_G rather than a ΔV_{th} ; (ii) the fast negative shift in V_{th} followed by a gradual recovery is attributable to bias stress, interface trapping, and mechanical/thermal relaxation; and (iii) chemical selectivity is unsubstantiated because

comparative data for other thiols were not presented. We rebut these claims by combining first-principles analysis with experiments. Specifically, we reexamine the EIFET architecture and measurement protocol, analyze the co-evolution of $V_{th}(t)$ with the IE surface energy $\gamma_s(t)$, and marshal the complete set of controls reported in the original article. This integrated analysis shows that the IE operates as a floating node (making a persistent IE–gate leakage path implausible) and that trap-only scenarios cannot account for the observed recovery kinetics or the dielectric-independent response. In what follows, we systematically refute each of the Comment's claims on these grounds.

2. Clarifying the EIFET architecture and measurement conditions

EIFET modifies an OCMFET-like stack by applying V_G to the gate (G) while using a floating IE (no external bias) as the chemically active surface separated from G by a dielectric (600 nm-thick Parylene C or 300 nm-thick SiO₂). Transfer curves are taken quasi-statically in saturation to extract V_{th} and μ_{FET} ; the IE is confined by a PDMS bank during droplet exposure

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(Fig. 1a and b in the original article¹). In this configuration, a surface reaction at the IE can modulate the effective gate bias at the channel *via* capacitive coupling, even though the IE is electrically isolated. Operationally, the IE is unbiased and the gate dielectric is designed to suppress dc leakage. If a sizable ohmic leakage path existed between IE and G, the device would collapse into an OCMFET-like conduction path instead of the reproducible, reversible V_{th} modulation observed. This point—central to our measurement geometry—was already emphasized in our internal rebuttal notes.

3. I_G vs. V_{th} shift: a capacitive-network view

The Comment correctly states that charge accumulation at the interference electrode (IE) is, in principle, accompanied by a transient displacement current, $I_G = \partial Q/\partial t$. We clarify, however, that in the operational context of the EIFET, I_G is not expected to be a meaningful sensing observable, because the IE is configured as a highly insulated, nominally floating node, and the gate dielectric's primary function is to suppress dc (ohmic) leakage. Accordingly, the experimentally observed response is governed primarily by accumulated IE charge (Q_{IE}), rather than by a sustained gate-current pathway. In a three-node network (G–dielectric–IE–channel fringe), a change in IE charge (ΔQ_{IE}) produces an IE potential shift $\Delta V_{\text{IE}} \approx \Delta Q_{\text{IE}}/C_{\Sigma}$, which partially offsets the applied V_G at the channel by a geometry-dependent coupling factor (β), yielding $V_{\text{th}} \rightarrow V_{\text{th}} + \beta\Delta V_{\text{IE}}$. This ΔV_{th} modulation is therefore a quasi-static effect arising from ΔQ_{IE} , which constitutes the primary sensing signal.

Quantitatively, the measured gate-dielectric areal capacitances are $C_1 = 3.70 \pm 0.2 \text{ nF cm}^{-2}$ (Parylene C) and $10.8 \pm 0.1 \text{ nF cm}^{-2}$ (300 nm SiO₂). A representative shift $|\Delta V_{\text{th}}| \sim 10 \text{ V}$ (from +2.2 to –7.6 V within ~ 15 min in Fig. 1d and 2) corresponds to an equivalent areal charge density $\Delta Q/A \approx C_1\Delta V \sim 3.7 \times 10^{-8} - 1.1 \times 10^{-7} \text{ C cm}^{-2}$, *i.e.*, $\sim 2.3 \times 10^{11} - 6.7 \times 10^{11} \text{ e cm}^{-2}$. Only a small fraction of a thiol monolayer's electron inventory would be required to generate this modulation, and thus the proposed mechanism does not rely on a large or prolonged I_G .

The Comment estimates an average $I_G \sim 10 \text{ nA cm}^{-2}$ over 2 hr by assuming full monolayer coverage with a 0.3 nm^2 footprint; we clarify that this estimate effectively reflects an upper-bound scenario based on assumptions not appropriate for a nominally floating IE configuration. In particular, it implicitly (i) adopts monolayer-scale charge generation, (ii) assumes near-quantitative and effectively instantaneous charge transfer, and (iii) treats the IE as if it were a driven electrode coupled to a charge reservoir. In our device, by contrast, the reacting IE area confined by the PDMS bank is mm^2 -scale and the IE remains nominally floating; therefore, charge accumulation and relaxation are governed by capacitive coupling and interfacial equilibration rather than by a forced current path. Under these conditions, any displacement-current component, if present, would be transient and small during quasi-static

transfer-curve acquisition. Importantly, the presence or absence of such a negligible transient I_G is secondary and does not invalidate the robust, reproducible quasi-static $V_{\text{th}}(t)$ dynamics driven by ΔQ_{IE} .

4. Controls that separate chemistry from bias-stress/artefacts

The instantaneous rate dV_{th}/dt and the IE surface energy $\gamma_s(t)$ evolve in opposite directions on the same time axis: a rapid early drop in γ_s (adsorption/ordering) coincides with the peak in $|dV_{\text{th}}/dt|$ within ~ 4 min (“Section I”), followed by a slower regime (“Section II”) and eventual saturation (“Section III”), exactly mirroring the V_{th} kinetics (Fig. 2a–c). This co-variation of an electrical observable with an independent surface-chemistry metric is difficult to reconcile with a trap-only mechanism. We raised the same questions raised by the authors and conducted additional comparative experiments. First, replacing Parylene C with 300 nm SiO₂ (identical channel material and geometry) reproduces the same qualitative $V_{\text{th}}(t)$ signature, including the fast negative shift and subsequent recovery as shown in Fig. 4c and d of the original paper. If polymer infiltration/swelling or a particular trap landscape in Parylene C were dominant, the response should depend strongly on the dielectric; it does not. Secondly, when the IE is pre-covered with a TP SAM, the V_{th} variation is negligible for 2 h under an identical protocol (Fig. 4e–g)—a decisive control, because the absence of further surface reaction suppresses the electrical signature. A trap-only explanation would not predict this loss of signal. Finally, long-term operation without IE exposure or exposure to non-polar toluene produces only minor drifts; ionic ammonium hydroxide and polar water show modest shifts with slight trends, but never reproduce the large, reversible pattern seen with thiols (Fig. 3). These data collectively rule out simple thermal/mechanical effects and generic bias-stress as the origin of the thiol response.

As a result, these same distinctions—dielectric-independence, SAM pre-coverage, solvent/ionic controls—are precisely those highlighted in our internal rebuttal, which also points out that a persistent IE–G leakage sufficient to drive the effect would degrade device operation—contrary to observation.

5. On the “recovery” and its compatibility with Au–thiol chemistry

The Comment views the ~ 80 min recovery toward the initial V_{th} as incompatible with molecular adsorption, inferring bias-stress instead. Our interpretation distinguishes chemical state from transient IE charge and clarifies that a nominally floating IE is not an ideally isolated conductor under droplet exposure. Because the IE is an Au surface directly interfaced with the liquid phase, the most physically plausible relaxation pathway is interfacial equilibration at the metal–solution boundary: non-equilibrium charge generated during the early stages of Au–thiol chemistry (adsorption on oxidized Au, reduction/thiolate



formation, H₂ evolution) can transiently bias the IE potential, yet subsequently relax through electrical double-layer charging/discharging and partial charge exchange (back-transfer) with the adjacent solution as the reaction rate decreases. In parallel, charge redistribution within the floating conductor and its interfacial region can further reduce the potential component that is capacitively coupled to the channel, even if the chemisorbed SAM remains intact. Finally, while the gate dielectric is designed to suppress *dc* (ohmic) leakage and a sizable persistent IE–G leakage path would contradict the stable, reproducible transfer characteristics, a finite but very small leakage/displacement pathway through the dielectric stack and surrounding materials cannot be strictly excluded and would, at most, act as a secondary discharge route on the experimental timescale. In this picture, the SAM remains while the net IE charge (and thus its capacitive influence) relaxes, allowing V_{th} to return near its baseline—exactly the evolution depicted schematically and observed experimentally (Fig. 1). The key point is that chemistry can be irreversible while the electrical V_{th} variation temporarily induced by Au–thiol reaction is reversible, because the variation depends on transient electrostatics coupled to the IE rather than solely on the final SAM coverage.

6. Chemical specificity and data availability

For clarity, the main text in the original paper focused on TP. We clarify, however, that the corresponding datasets for DT and PFBT were not presented previously primarily to avoid repetitive display of substantively similar time-dependent signatures

and to maintain readability, rather than due to a lack of comparative evidence. (The newly added data is presented in this Reply as Fig. 1.) In Fig. 1, Fig. 1(a) and (c) show the evolution of the $I_D^{0.5}$ – V_G characteristics during the thiol reaction on the IE, and Fig. 1(b) and (d) summarize the corresponding V_{th} values extracted from these transfer plots together with dV_{th}/dt as a function of reaction time for DT and PFBT, respectively. Importantly, TP, DT, and PFBT all exhibit the same overall kinetic outline—an initial rapid response followed by a slower regime and eventual saturation—supporting that the EIFET signal is a general feature of Au–thiol interfacial chemistry coupled to the IE rather than a molecule-specific artefact. At the same time, the characteristic timescales and early-time slopes differ across molecules in a chemically plausible manner: DT shows a prolonged evolution before saturation (rapid regime to ~ 7.5 min followed by a slower regime extending to ~ 44 min), whereas PFBT reaches saturation earlier (~ 24 min) while exhibiting a smaller initial $|dV_{th}/dt|$ than TP or DT, indicating a slower initial interfacial progression. This attenuated early-time response for PFBT may plausibly originate from the strongly fluorinated aromatic framework, which can modulate the electronic structure and interfacial energetics of the thiol motif; fluorine substitution may stabilize reactive intermediates and redistribute electron density over the ring *via* resonance/inductive effects, potentially reducing the effective driving force for the earliest surface-reaction steps. While these transient features should not be interpreted as definitive intrinsic kinetic constants, the consistent mechanistic signature across distinct thiol structures, together with systematic molecule-dependent timescales, directly addresses the concern regarding chemical specificity.

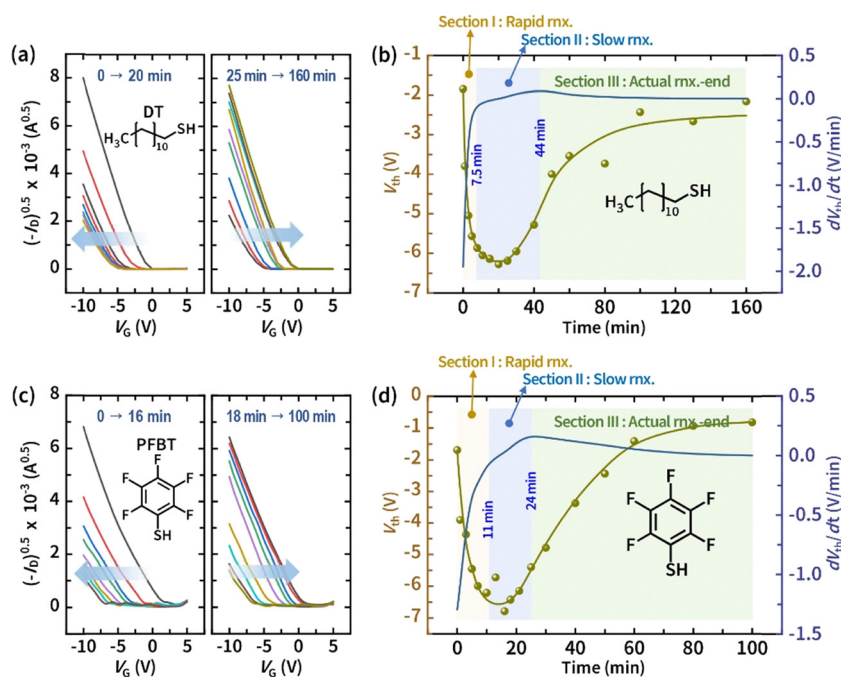


Fig. 1 (a) and (c) Variations of the $I_D^{0.5}$ – V_G plots of the EIFET device with progression of the thiol reaction on the IE surface for DT and PFBT, respectively. (b) and (d) Variations of V_{th} extracted from the corresponding $I_D^{0.5}$ – V_G plot and dV_{th}/dt according to the reaction time for DT and PFBT, respectively.



7. Quantitative consistency of the observed ΔV_{th}

Using the measured C_i , a $|\Delta V_{\text{th}}| \sim 10$ V shift corresponds to $\sim(2.3\text{--}6.7) \times 10^{11}$ e cm^{-2} . A full thiol monolayer contains $\sim 10^{14}$ molecules cm^{-2} ; thus, an extremely small fraction of the electrons transiently produced during the surface reaction need be retained in the IE to account for the observed ΔV_{th} . This scale analysis refutes the premise that a large sustained I_G must accompany the signal and aligns with a short-lived displacement current plus quasi-static capacitive rebiasing of the channel.

8. Conclusions

The EIFET response is best understood as a capacitive rebiasing of the channel by a transient IE potential that arises during the Au–thiol surface reaction. This mechanism explains (i) the distinctive $V_{\text{th}}(t)$ with fast onset and gradual recovery, (ii) its tight correlation with $\gamma_s(t)$, (iii) its robustness across different gate dielectrics, and (iv) its suppression by preformed SAMs. The trap-only/bias-stress hypothesis offered in the Comment does not reproduce this full pattern, and the overall body of evidence strongly supports the interpretation that the observed V_{th} modulation is directly linked to the Au–thiol surface reaction at the interference electrode.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

Fig. 1 were added for this Reply article. All remaining data supporting the findings and arguments presented here are available within the original published article (*J. Mater. Chem. C*, 2021, **9**, 8179–8188) and its supplementary information (SI).

Any additional information relevant to this Reply is entirely derived from those previously published sources.

Acknowledgements

We thank Micjan and Weis for their constructive critique and for stimulating this clarification of mechanism and controls. Please note that the figure numbers used in this rebuttal follow those in the original paper; Fig. 1 has been added in this Reply to present previously collected data not included in the original article/SI.

References

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