Note S6 | QAOA details (Ising mapping, γ– β search, and sampling) for MXene EIS fitting

The discrete quantum branch uses QAOA to explore a quantized parameter space for the MXene EIS circuit while retaining physical feasibility through bounded decoding. The overall workflow is: (i) construct a *local quadratic surrogate* of the classical complex-domain objective around a baseline parameter set, (ii) discretize seven parameters using a fixed bit depth (3 bits/parameter), (iii) convert the resulting QUBO into an Ising Hamiltonian $H(\boldsymbol{z})$, (iv) select near-optimal QAOA angles (γ, β) by a two-stage grid search, and (v) sample the optimized QAOA circuit with shots = 4096 and decode the best observed bitstring into θ for EIS overlay.

S6.1 From EIS objective to a local quadratic surrogate.

The fitting objective is the complex-domain sum of squares,

$$F(\theta) = \sum_{k=1}^{N} |Z_{model}(\omega_k; \theta) - Z_{data}(\omega_k; \theta)|^2 \quad \text{(S22)}$$

Because QAOA requires a discrete optimization target, we approximate FFF locally in a bounded coordinate $u \in [0,1]^7$ (**Note S3**). Let u_0 be the baseline point (classical fit) and define $\delta u = u - u_0$. A second-order expansion yield

$$\hat{F}(u) = F(u_0) + \mathbf{g}^{\top} \delta u + \frac{1}{2} \delta u^{\top} \mathbf{H}\, \delta u \quad \text{(S23)}$$

where $\mathbf{g}$ and $\mathbf{H}$ are the gradient and Hessian in u-space, estimated via finite differences. This quadratic surrogate captures the dominant local curvature and parameter couplings and is sufficiently structured to convert into a binary quadratic form.

S6.2 3-bit discretization and trust-region decoding.

Each parameter is encoded by 3 bits (b_{i0}, b_{i1}, b_{i1}), giving 8 discrete levels. We convert the 3-bit integer $k_i = b_{i0} + 2b_{i1} + 4b_{i2} \in \{0, \ldots, 7\}$ into a normalized level $s_i = k_i/7 \in [0,1]$. To keep the surrogate valid and prevent large, unphysical jumps, we restrict the discrete search to a *local trust region* of width Δ around u_0:

$$u_i = clip\,(u_{0,i} + \Delta\,(2s_i - 1), 0, 1) \tag{S24}$$

with $\Delta = 0.08$ in this study. The decoded u is then mapped to physical parameters θ using the bound rules of **Table S1** (linear/log decoding). This makes the discrete QAOA output directly comparable to the continuous VQE/VQA and classical results.

S6.3 QUBO construction and Ising mapping.

Substituting the discretized $u(b)$ into $\hat{F}(u)$ yields a quadratic polynomial in the binary variables:

$$\hat{F}(u) = \mathbf{b}^{\top}\mathbf{Q}\mathbf{b} + \mathbf{q}^{\top}\mathbf{b} + const. \tag{S25}$$

This is a standard QUBO. To run QAOA, we convert it to an Ising form over spins $z_i \in \{-1, +1\}$ using $b_i = (1 - z_i)/2$. The resulting Hamiltonian is

$$H(z) = \sum_i h_i z_i + \sum_{i<j} J_{ij} z_i z_j + const \tag{S26}$$

where h_i and J_{ij} are computed analytically from $\mathbf{Q}$ and $\mathbf{q}$. In this work the Ising instance has 21 spins (3 bits × 7 parameters) and is fully specified by the exported coefficient lists (h_i, J_{ij}).

S6.4 QAOA circuit, γ–β search, and refinement.

We use QAOA depth $p = 1$ to maintain low circuit depth while capturing non-trivial correlations. The variational state is

$$|\,\psi(\gamma, \beta)\rangle = e^{-i\beta \sum_i X_i} e^{-i\gamma H(\boldsymbol{Z})} |+\rangle^{\otimes 21} \tag{S27}$$

where $H(\boldsymbol{Z})$ is diagonal in the computational basis and X_i is the Pauli-X mixer. The expected energy is

$$E(\gamma, \beta) = \langle \psi(\gamma, \beta) \mid H \mid \psi(\gamma, \beta) \rangle \tag{S28}$$

Angles are selected by a two-stage grid strategy: a coarse (γ, β) sweep over a 5×5 grid to locate the approximate basin, followed by a refined 9×9 window centered at the best coarse cell to sharpen the optimum. The final chosen (γ^*, β^*) is the minimum-energy grid point in the refined window and is reported along with the corresponding heatmaps (**Fig. S6a-b**).

S6.5 Sampling (shots = 4096) and best-shot decoding.

With (γ^*, β^*) fixed, the QAOA circuit is sampled with 4096 shots. Each measured bitstring corresponds to a discrete parameter level vector s, which is decoded to u using the trust-region rule above and then to θ via Table S1. We report the top-10 bitstring counts to assess concentration and multimodality; the "best-shot" solution is defined as the sampled bitstring with the lowest Ising energy and its decoded θ is used for the final Nyquist/Bode overlays (**Fig. 3**) and the SI summary table of decoded parameters and bitstring frequencies (**Fig. S6**). This procedure mirrors realistic hardware operation, where one obtains a distribution of candidate solutions from measurement rather than a single deterministic optimum.