Supplemental Material

Emergent superconductivity in doped ferroelectric hafnia

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I. SWITCHING BARRIER

The switching barrier as a function of electron-doping concentration (n_e) in $Pca2_1$ HfO_2 is shown in Fig. S1.The barrier height in undoped HfO_2 ($n_e = 0.0$) is 0.33 eV per formula unit (eV/f.u.), which is in good agreement with previous reports [1]. It is evident that the switching barrier depends on n_e weakly.

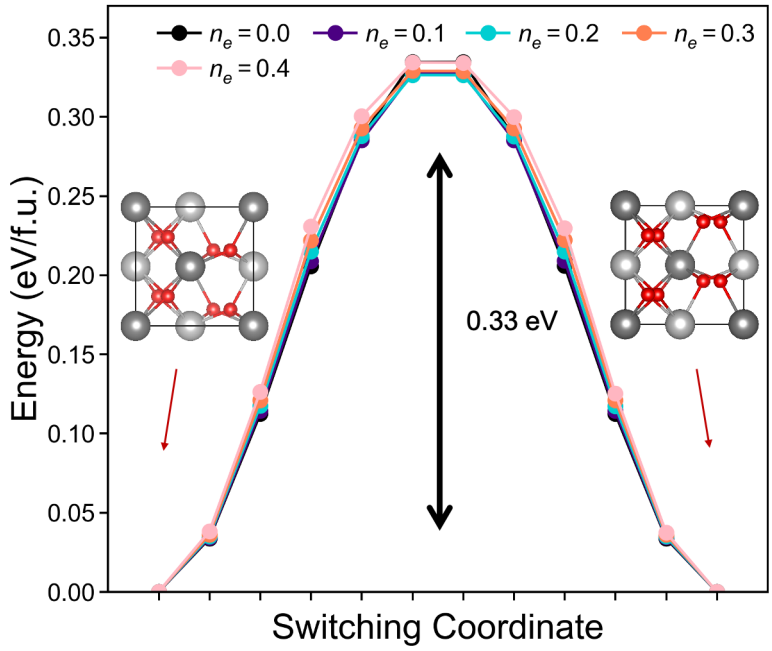

FIG. S1. Switching barrier of $Pca2_1$ HfO_2 as a function of electron-doping concentration (n_e).

II. ELIASHBERG SPECTRAL FUNCTION AND ELECTRON-PHONON COUPLING

The isotropic Eliashberg spectral function $\alpha^2F(\omega)$ and mode-resolved electron-phonon coupling strength $\lambda_{\mathbf{q}\nu}$ are calculated using the EPW code [2–4]. The imaginary part of the phonon self-energy is expressed within the double-delta approximation as

$$\mathrm{Im}\Pi_{\mathbf{q}\nu} = 2\pi\omega_{\mathbf{q}\nu}\sum_{ij}\int\frac{d\mathbf{k}}{V_{\mathrm{BZ}}}|g^{\nu}_{ij}(\mathbf{k},\mathbf{q})|^2\delta(\epsilon_{j\mathbf{k}}-\epsilon_F)\delta(\epsilon_{i\mathbf{k}+\mathbf{q}}-\epsilon_F). \tag{1}$$

Here, $\omega_{\mathbf{q}\nu}$ and V_{BZ} is the phonon frequency of νth phonon mode at wave vector $\mathbf{q}$ and the volume of Brillouin zone(BZ), respectively; ϵ_F is the Fermi energy. The electron-phonon matrix elements, $g^{\nu}_{ij}(\mathbf{k},\mathbf{q})$, characterizing the scattering process between Kohn-Sham states $i\mathbf{k}+\mathbf{q}$ and j$\mathbf{k}$, is given as

$$g^{\nu}_{ij}(\mathbf{k},\mathbf{q}) = \frac{1}{\sqrt{2\omega_{\mathbf{q}\nu}}}\langle\psi_{i\mathbf{k}+\mathbf{q}}\,|\partial_{\mathbf{q}v}V|\psi_{j\mathbf{k}}\rangle, \tag{2}$$

where $\partial_{\mathbf{q}v}V$ is the derivative of the self-consistent potential associated with the phonon mode $\mathbf{q}\nu$, and $\psi_{j\mathbf{k}}$ is the electronic wavefunction for band j and wavevector $\mathbf{k}$. The mode-resolved electron-phonon coupling strength $\lambda_{\mathbf{q}\nu}$ can be related to the $\mathrm{Im}\Pi_{\mathbf{q}\nu}$ as

$$\lambda_{\mathbf{q}\nu} = \frac{1}{\pi N_F}\frac{\mathrm{Im}\Pi_{\mathbf{q}\nu}}{\omega^2_{\mathbf{q}\nu}} = \frac{1}{N_F\omega_{\mathbf{q}\nu}}\sum_{ij}\int\frac{d\mathbf{k}}{V_{\mathrm{BZ}}}|g^{\nu}_{ij}(\mathbf{k},\mathbf{q})|^2\delta(\epsilon_{j\mathbf{k}}-\epsilon_F)\delta(\epsilon_{i\mathbf{k}+\mathbf{q}}-\epsilon_F), \tag{3}$$

where N_F is the density of states per spin at the Fermi level. The isotropic Eliashberg spectral function $\alpha^2F(\omega)$ can be obtained through a BZ integration of $\lambda_{\mathbf{q}\nu}$ as

$$\alpha^2F(\omega) = \frac{1}{2}\sum_{\nu}\int\frac{d\mathbf{q}}{V_{\mathrm{BZ}}}\omega_{\mathbf{q}\nu}\lambda_{\mathbf{q}\nu}\delta(\omega-\omega_{\mathbf{q}\nu}). \tag{4}$$

The accumulative electron-phonon coupling $\lambda(\omega)$ is defined as

$$\lambda(\omega) = 2\int_0^{\omega}\frac{\alpha^2F(\omega)}{\omega}d\omega \tag{5}$$

and the total electron-phonon coupling λ is defined as:

$$\lambda = 2\int_0^{\infty}\frac{\alpha^2F(\omega)}{\omega}d\omega \tag{6}$$

We calculate the isotropic Eliashberg spectral function $\alpha^2F(\omega)$ and accumulative electron-phonon coupling strength $\lambda(\omega)$ for $Pca2_1$ HfO_2 with different values of n_e, as shown in Fig. S2. As n_e increases, the peaks of $\alpha^2F(\omega)$ experience both red-shifting and amplification. Correspondingly, there is an increment in $\lambda(\omega)$ associated with increasing n_e, which subsequently leads to an enhancement of superconducting transition temperature T_c.

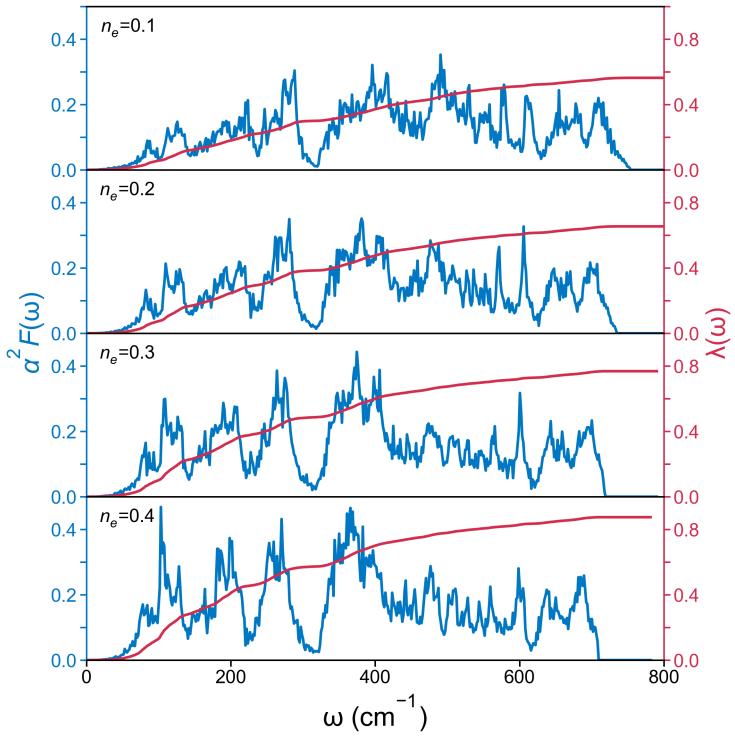

FIG. S2. Isotropic Eliashberg spectral function $\alpha^2F(\omega)$ (blue line) and accumulative electron-phonon coupling strength $\lambda(\omega)$ (red line) at various values of n_e.

III. COULOMB PSEUDOPOTENTIAL μ^* AND SUPERCONDUCTING TRANSITION TEMPERATURE T_c

Coulomb pseudopotential μ^* is an empirical parameter which describes the effective screened Coulomb repulsion and can strongly affect the estimate of superconducting T_c. We calculate T_c as a function of μ^* at $n_e = 0.3$ using two methods, the McMillan-Allen-Dynes formula (T_c^{MAD}) [5] and the solution of the anisotropic Eliashberg equations ($T_c^{\mathrm{Elia.}}$) [2, 3]. As shown in Fig. S3, our test reveals that $T_c^{\mathrm{Elia.}}$ decreases from ≈31 K to ≈4 K while T_c^{MAD} changes from ≈24 K to ≈0.9 K, when μ^* increases from 0 to 0.3. These values remain comparable with those of known superconducting binary oxides. For results reported in the main text, $\mu^* = 0.1$ is used.

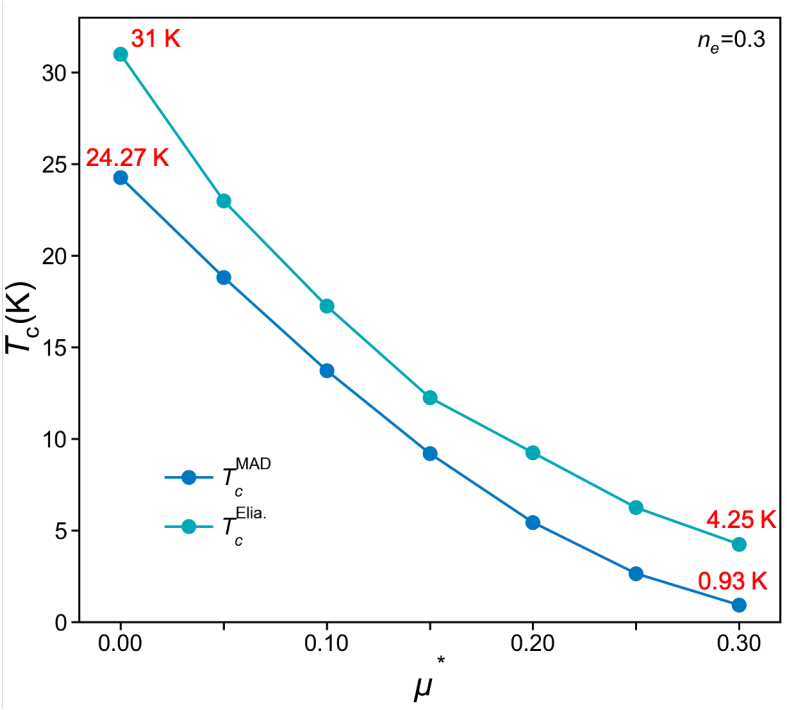

FIG. S3. Superconducting transition temperature T_c estimated by the McMillan-Allen-Dynes formula and the solution of Eliashberg equations as a function of μ^*.

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