Supplementary Material: Nonlinear Nanocircuitry Based on Quantum Tunneling Effects

S1. Tunneling-Induced Quantum Conductance

The quantum conductance yielded by the tunneling effect has both linear and nonlinear components. The linear, in-phase response of the current at the fundamental frequency (m=1) may lead to the power dissipation, and the quantum conductance is given by [1]-[2]

$$G_{\omega}^{(1)} = \frac{q}{\hbar\omega}\frac{I_{\text{dark}}\left(V_{\text{dc}}+\hbar\omega/q\right)-I_{\text{dark}}\left(V_{\text{dc}}-\hbar\omega/q\right)}{2}, \tag{S1}$$

Although nonlinear terms in Eq. (3) in the main text are small, they can have relevant effects when the local field strength is strong. In the nonlinear regime, there exists high-order dissipative and harmonic contributions. The higher-order terms can derived by extending the expansion in Eq. (2) in the main text. The third and other high-order quantum conductance, related to the multi-photon absorption, are derived as:

$$G_{\omega}^{(3)} = \left(\frac{q}{2\hbar\omega}\right)^{3}\left\{\begin{array}{l} -I_{\text{dark}}\left(V_{\text{dc}}+\hbar\omega/q\right)+I_{\text{dark}}\left(V_{\text{dc}}-\hbar\omega/q\right) \\ +\frac{1}{2}\left[I_{\text{dark}}\left(V_{\text{dc}}+2\hbar\omega/q\right)-I_{\text{dark}}\left(V_{\text{dc}}-2\hbar\omega/q\right)\right] \end{array}\right\}. \tag{S2}$$

$$G_{\omega}^{(5)} = \left(\frac{q}{2\hbar\omega}\right)^{5}\left\{\begin{array}{l} \frac{5}{12}\left[I_{\text{dark}}\left(V_{\text{dc}}+\hbar\omega/q\right)-I_{\text{dark}}\left(V_{\text{dc}}-\hbar\omega/q\right)\right] \\ -\frac{1}{3}\left[I_{\text{dark}}\left(V_{\text{dc}}+2\hbar\omega/q\right)-I_{\text{dark}}\left(V_{\text{dc}}-2\hbar\omega/q\right)\right] \\ +\frac{1}{12}\left[I_{\text{dark}}\left(V_{\text{dc}}+3\hbar\omega/q\right)-I_{\text{dark}}\left(V_{\text{dc}}-3\hbar\omega/q\right)\right] \end{array}\right\}. \tag{S3}$$

$$G_{\omega}^{(7)}=\left(\frac{q}{2\hbar\omega}\right)^{7}\left\{\begin{array}{l}-\frac{7}{72}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-\hbar\omega/q\right)\right]\\+\frac{7}{72}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+2\hbar\omega/q\right)-I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-2\hbar\omega/q\right)\right]\\-\frac{1}{24}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+3\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-3\hbar\omega/q\right)\right]\\+\frac{1}{144}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+4\hbar\omega/q\right)-I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-4\hbar\omega/q\right)\right]\end{array}\right\}. \quad \text{(S4)}$$

The nonlinear quantum conductivities related to the second-harmonic generation can be derived as:

$$G_{2\omega}^{(2)}=\left(\frac{q}{2\hbar\omega}\right)^{2}\left\{\begin{array}{l}I_{\mathrm{dark}}\left(V_{\mathrm{dc}}\right)-\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-\hbar\omega/q\right)\right]\\+\frac{1}{2}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+2\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-2\hbar\omega/q\right)\right]\end{array}\right\} \text{(S5)}$$

$$G_{2\omega}^{(4)}=\left(\frac{q}{2\hbar\omega}\right)^{4}\left\{\begin{array}{l}-\frac{4}{3}I_{\mathrm{dark}}\left(V_{\mathrm{dc}}\right)+\frac{7}{6}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-\hbar\omega/q\right)\right]+\\-\frac{2}{3}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+2\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-2\hbar\omega/q\right)\right]\\+\frac{1}{6}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+3\hbar\omega/q\right)+I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-3\hbar\omega/q\right)\right]+\end{array}\right\} \text{(S6)}$$

$$G_{2\omega}^{(6)}=\left(\frac{q}{2\hbar\omega}\right)^{6}\left\{\begin{array}{l}\frac{5}{8}I_{\mathrm{dark}}\left(V_{\mathrm{dc}}\right)-\frac{13}{24}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+\hbar\omega/q\right)-I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-\hbar\omega/q\right)\right]+\\+\frac{1}{8}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+2\hbar\omega/q\right)-I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-2\hbar\omega/q\right)\right]\\-\frac{1}{3}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+3\hbar\omega/q\right)-I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-3\hbar\omega/q\right)\right]\\+\frac{1}{48}\left[I_{\mathrm{dark}}\left(V_{\mathrm{dc}}+4\hbar\omega/q\right)-I_{\mathrm{dark}}\left(V_{\mathrm{dc}}-4\hbar\omega/q\right)\right]\end{array}\right\} \text{(S7)}$$

The second harmonic generation and the intensity-dependent (resistive) self-modulation are mainly contributed by the lowest-order approximation term $G_{2\omega}^{(2)}$ and $G_{\omega}^{(3)}$.

S2. Static Tunneling Current in a MOM Junction

According to the Simmon's model [3], when a dc bias V_{dc} is applied to the MOM junction, the tunneling current density can be expressed as:

$$J_{\mathrm{dark}}\left(V_{\mathrm{dc}}\right)=\frac{m_0 q}{2\pi^2\hbar^3}\int_0^{\infty}D\left(\mathscr{E}_x\right)d\mathscr{E}_x\int_{\mathscr{E}_x}^{\infty}\left[F_1\left(\mathscr{E}\right)-F_2\left(\mathscr{E}+qV_{\mathrm{dc}}\right)\right]d\mathscr{E}, \qquad \text{(S8)}$$

where q is the electron charge, m_0 is the electron mass, $\hbar$ is the reduced Plank constant, F_i is the Fermi-Dirac distribution function of the *i*-th metal, $\mathscr{E}$ and $\mathscr{E}_x$ are respectively the total and longitudinal energy of electrons, D is the electron transmission probability. The charge transport mechanism in the MOM junction depends on the difference between Fermi levels of two metals. When a dc bias is applied, currents can flow between two metals by means of the tunneling effect. At equilibrium, the well-known Simmons formula can describe the current-voltage behavior of this MOM junction [3]:

$$J_{\mathrm{dark}}\left(V_{\mathrm{dc}}\right)=J_{1\to 2}-J_{2\to 1}=J_{\mathrm{dark},0}\left[\bar{\varphi}\exp\left(-A\sqrt{\bar{\varphi}}\right)-\left(\bar{\varphi}+qV_{\mathrm{dc}}\right)\exp\left(-A\sqrt{\bar{\varphi}+qV_{\mathrm{dc}}}\right)\right] \qquad \text{(S9)}$$

where $J_{\mathrm{dark},0}=q/2\pi h\left(\beta\Delta s\right)^2$, $J_{1\to 2}=J_{\mathrm{dark},0}\left[\bar{\varphi}\exp\left(-A\sqrt{\bar{\varphi}}\right)\right]$ is the current density flowing from metal 1 to metal 2, and $J_{2\to 1}=J_{\mathrm{dark},0}\left(\bar{\varphi}+qV_{\mathrm{dc}}\right)\exp\left(-A\sqrt{\bar{\varphi}+qV_{\mathrm{dc}}}\right)$ is

the current density flowing from metal 2 to metal 1, $A = 4\sqrt{2m_0}\pi\beta\Delta s / h$, β is the correction factor defined in [S3] as $\beta = 1 - [1/(8\bar{\varphi}^2 \Delta s)]\int_{s_1}^{s_2} [\varphi(x) - \bar{\Phi}]^2 \, dx$ (s_1 and s_2 are limits of barrier at Fermi level), $\bar{\varphi}$ and $\Delta s = s_2 - s_1$ are the mean potential barrier height relative to Fermi level and barrier width for tunneling electrons.

In the reverse bias mode, of which the metal 1 with lower work function is negatively biased, the height of tunneling barrier between two metals, which takes into account the potential barrier lowering due to the image force, is given by

$$\begin{aligned} \varphi(x) &= \varphi_1 + \left(\Delta\varphi - qV\right)\frac{x}{s} - \frac{q^2}{4\pi K\varepsilon_0 s}\left[\frac{x}{2} + \sum_{n=1}^{\infty}\frac{ns}{\left[\left(ns\right)^2 - x^2\right]} - \frac{1}{ns}\right] \\ &\approx \varphi_1 + \left(\Delta\varphi - qV\right)\frac{x}{s} - 1.15\ln[2]\frac{q^2}{8\pi K\varepsilon_0}\frac{s}{x(s-x)}, \end{aligned} \tag{S10}$$

where $\Delta\varphi = \varphi_2 - \varphi_1$, $\varphi_i = \Phi_i - \chi$ is the barrier heights at the interface between metal $\underline{i}$ and oxide, Φ_i is the work function of metal i, and χ is the electron affinity of oxide. From [S3], the mean barrier height $\bar{\varphi}$ is given by:

$$\bar{\varphi} = \frac{1}{\Delta s}\int_{s_1}^{s_2}\varphi(x)dx \approx \varphi_1 + \frac{s_2 + s_1}{2s}(\Delta\varphi - qV) - 1.15\ln\left[2\right]\frac{q^2}{8\pi K\varepsilon_0\Delta s}\ln\left[\frac{s_2(s-s_1)}{s_1(s-s_2)}\right] \tag{S11}$$

where s is the barrier height and limits of barrier at Fermi level s_1 and s_2 are given by the real roots of the cubic equation:

$$\varphi_1 + \left(\Delta\varphi - qV\right)\frac{x}{s} - 1.15\ln\left[2\right]\frac{q^2}{8\pi K\varepsilon_0}\frac{s}{x(s-x)}. \tag{S12}$$

From (S9) and (11), the reverse-bias dark current density can be calculated.

In the forward bias mode, of which the metal 1 with lower work function is positively biased, the mean barrier height $\bar{\varphi}$ is given by:

$$\bar{\varphi} \approx \varphi_2 - \frac{s_2 + s_1}{2s}(\Delta\varphi + qV) - 1.15\ln[2]\frac{q^2}{8\pi K\varepsilon_0 \Delta s}\ln\left[\frac{s_2(s-s_1)}{s_1(s-s_2)}\right] \qquad \text{(S13)}$$

where s_1 and s_2 are given by the real roots of the cubic equation:

$$\varphi_2 + (\Delta\varphi + qV)\frac{x}{s} - 1.15\ln[2]\frac{q^2}{8\pi K\varepsilon_0}\frac{s}{x(s-x)}. \qquad \text{(S14)}$$

From (S9) and (S13), the forward-bias dark current density can be calculated. In a MOM structure, a transition to hopping or diffusive transport is difficult, since the field-induced breakdown occurs prior to reaching the bias necessary to bring about the change in mechanism, typical for quantum tunneling devices. When the applied bias is increased from zero to a high voltage, the carrier transport mechanism would transit from direct tunneling to field emission (Fowler Nordheim tunneling).

References

[1] P. K. Tien and J. P. Gordon, *Phy. Rev*. **129**, 647 (1963).

[2] J .R. Truker, *IEEE J. Quantum Electron.* **15**, 1234 (1979).

[3] J. G. Simmons, *J. Appl. Phys.*, **34**, 1793 (1963); J. G. Simmons, *J. Appl. Phys.* **34**, 2581 (1963); J .G. Simmons, *J. Appl. Phys*. **35**, 2655 (1964).