Supporting Information: Ballistic-like space-charge-limited currents in halide perovskites at room temperature

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S1. Potential-dependent drift velocity

Assuming φ is the electrostatic potential of a sample of thickness L, where $\varphi(0) = 0$ and $\varphi(L) = V$, being V the external applied voltage, then the drift velocity can be taken as

$$v_d = k\varphi^p \tag{S1}$$

where p is a dimensionless constant and k a constant whose units depend on p. Considering the current density J independent of the position x and φ, it can be expressed as

$$J = Q\ N\ v_d \tag{S2}$$

where Q and N are the charge and concentration of the charge carrier, respectively. By substituting Equation (S1) in (S2) one obtains

$$N = \frac{J}{Qk}\ \varphi^{-p} \tag{S3}$$

Substituting Equation (S3) in the Poisson equation it results in

$$\frac{d^2\varphi}{dx^2} = \frac{K}{\varphi^p} \tag{S4}$$

where $K = J(k\epsilon_0\epsilon_r)^{-1}$. Then, we multiply Equation (S4) by $2d\varphi = 2(d\varphi/dx)dx$

$$2\frac{d\varphi}{dx}\left(\frac{d^2\varphi}{dx^2}dx\right) = 2\frac{K}{\varphi^p}\ d\varphi \tag{S5}$$

Subsequently, by integrating Equation (S5) we obtain

$$\left(\frac{d\varphi}{dx}\right)^2 - \left(\frac{d\varphi}{dx}\right)_0^2 = \frac{2K}{1-p}\ \varphi^{1-p} \tag{S6}$$

Neglecting the second term of the left member and elevating at the power of ½ Equation (S6), we obtain

$$\frac{d\varphi}{dx} = \left(\frac{2K}{1-p}\right)^{1/2}\ \varphi^{\frac{1-p}{2}} \tag{S7}$$

Equation (S7) can be reordered for integration between 0 and x, where $\varphi(0) = 0$ and $\varphi(x) = \varphi$, respectively, then

$$2\frac{\varphi^{\frac{p+1}{2}}}{(p+1)} = \left(\frac{2K}{1-p}\right)^{1/2} x \tag{S8}$$

Equation (S8) can be reordered and elevated at the power of $2(p+1)^{-1}$ to obtain the generic electrostatic potential as

$$\varphi = (p+1)^{\frac{2}{p+1}} \left(\frac{J}{2k\epsilon_0\epsilon_r(1-p)} \right)^{\frac{1}{p+1}} x^{\frac{2}{p+1}} \tag{S9}$$

Equation (S8) can also be reordered, elevated at the power of 2 and evaluated at $x = L$, where $\varphi(L) = V$, to obtain the generic current density

$$J = \kappa \frac{V^{p+1}}{L^2} \tag{S10}$$

where the constant κ is

$$\kappa = 2k\epsilon_0\epsilon_r \frac{(1-p)}{(p+1)^2} \tag{S11}$$

In the ballistic regime, the Child-Langmuir law[1, 2] of space-charge-limited current (SCLC) uses $k = (2Q/M)^{1/2}$ and $p = 1/2$, then the electrostatic potential results as

$$\varphi = \left(\frac{3}{2}\right)^{\frac{4}{3}} \left(\frac{J}{\epsilon_0\epsilon_r} \sqrt{\frac{M}{2Q}} \right)^{\frac{2}{3}} x^{\frac{4}{3}} \tag{S12}$$

and the current density is

$$J = \frac{4\epsilon_0\epsilon_r\mu_0}{9L^2} \sqrt{\frac{2Q}{M}}\, V^{3/2} \tag{S13}$$

The ballistic current density as in Equation (S13) is illustrated in Figure S1 for typical ranges of sample thickness and applied external voltage.

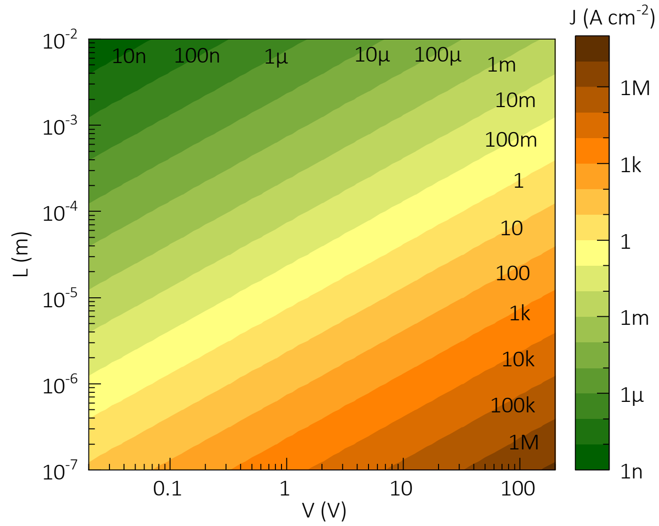

Figure S1: Current density as a function of the external applied voltage for the ballistic Child-Langmuir law[1, 2] of SCLC for a $MAPbI_3$ sample with ϵ_r=23[3] and Q and M being the electron charge and mass, respectively, in Equation (S13).

In the quasi-ballistic velocity-dependent dissipation (QvD)[4] regime of SCLC, $k = \left(\sqrt{2+\beta} - \sqrt{\beta}\right)\sqrt{\frac{Q}{M}}$ and $p = 1/2$, then the electrostatic potential results as

$$\varphi = \left(\frac{3}{2}\right)^{\frac{4}{3}} \left(\frac{J}{\epsilon_0 \epsilon_r} \sqrt{\frac{M}{Q}}\right)^{\frac{2}{3}} x^{\frac{4}{3}} \qquad \text{(S14)}$$

and the current density is

$$J = \frac{4\epsilon_0\epsilon_r\left(\sqrt{2+\beta} - \sqrt{\beta}\right)}{9L^2} \sqrt{\frac{Q}{M}}\, V^{3/2} \qquad \text{(S15)}$$

S2. Field-dependent drift velocity

Differently, the drift velocity can also be taken proportional to the absolute electric field $|\xi| = |d\varphi/dx|$, as

$$v_d = c\,\frac{d\varphi^p}{dx} \qquad \text{(S16)}$$

where c is a constant whose units depend on the dimensionless power p. Substituting Equation (S16) in (S2) one obtains the total charge carrier concentration as:

$$N = \frac{J}{Qk}\,\frac{d\varphi^{-p}}{dx} \qquad \text{(S17)}$$

Subsequently, substituting Equation (S17) in the Poisson equation it results in

$$\frac{d^2\varphi}{dx^2} = \frac{C}{\left(\frac{d\varphi}{dx}\right)^p} \qquad \text{(S18)}$$

where $C = J(c\,\epsilon_0\epsilon_r)^{-1}$. Then, we multiply Equation (S18) by $\left(\frac{d\varphi}{dx}\right)^p dx$

$$\left(\frac{d\varphi}{dx}\right)^p \frac{d^2\varphi}{dx^2}\,dx = C\,dx \qquad \text{(S19)}$$

Integrating Equation (S19) we obtain,

$$\left(\frac{d\varphi}{dx}\right)^{p+1} - \left(\frac{d\varphi}{dx}\right)_0^{p+1} = (p+1)Cx \qquad \text{(S20)}$$

Neglecting the second term of the left member and elevating Equation (S20) at the power of $(p+1)^{-1}$ we obtain

$$\frac{d\varphi}{dx} = \left((p+1)Cx\right)^{\frac{1}{p+1}} \tag{S21}$$

Equation (S21) can be reordered for integration between $x = 0$ and x, where $\varphi(0) = 0$ and $\varphi(x) = \varphi$, respectively, then the electrostatic potential can be found as

$$\varphi = \frac{(p+1)^{\frac{p+2}{p+1}}}{(p+2)} \left(\frac{J}{c\,\epsilon_0 \epsilon_r}\right)^{\frac{1}{p+1}} x^{\frac{p+2}{p+1}} \tag{S22}$$

Equation (S22) can be evaluated at $x = L$, where $\varphi(L) = V$, then the general current density results as

$$J = \varsigma \frac{V^{p+1}}{L^{p+2}} \tag{S23}$$

where the constant ς is

$$\varsigma = c\epsilon_0 \epsilon_r \frac{(p+2)^{p+1}}{(p+1)^{p+2}} \tag{S24}$$

In the Mott-Gurney law[5] of SCLC, $c = \mu$ and $p = 1$, therefore, the electrostatic potential results as

$$\varphi = \frac{2}{3}\sqrt{\frac{2J}{\mu\,\epsilon_0 \epsilon_r}}\, x^{\frac{3}{2}} \tag{S25}$$

and the current density is

$$J = \frac{9\epsilon_0 \epsilon_r \mu}{8L^3} V^2 \tag{S26}$$

The current as in Equation (S26)(S13) is illustrated in Figure S2 for typical values.

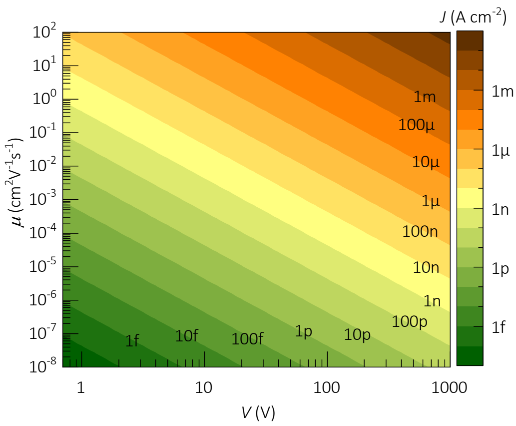

Figure S2: Current density as a function of the external applied voltage and mobility for the Mott-Gurney law[5] of mobility regime of SCLC for a $MAPbI_3$ sample with ϵ_r=23[3] and L=1.0 mm in Equation (S26)(S13).

In the ballistic-like voltage-dependent mobility (BVM) regime, $c = \mu_0 (V_0/L)^{1/2}$ and $p = 1/2$, then the electrostatic potential results as

$$\varphi = \frac{3}{5}\left(\frac{3}{2}\right)^{\frac{2}{3}}\left(\frac{J}{\epsilon_0 \epsilon_r \mu_0}\sqrt{\frac{L}{V_0}}\right)^{\frac{2}{3}} x^{\frac{5}{3}} \tag{S27}$$

and, considering that $\sqrt{500/243} \approx \sqrt{2}$, the current density can be approximated to

$$J = \frac{\epsilon_0 \epsilon_r \mu_0}{L^3}\sqrt{2V_0}V^{3/2} \tag{S28}$$

S2.1. The onset voltage V_0 of the BVM regime of SCLC

In the SCLC formalism, the $v_d \propto (d\varphi/dx)^{1/2}$ can explain a $J \propto V^{3/2}$, as above demonstrated. Typically, in the mobility regime the absolute value of the drift velocity is considered as

$$v_d = \mu \xi \tag{S29}$$

The use of Equation (S29) leads to the Mott-Gurney law.[5] However, assuming a transition from Ohmic to the BVM regime around an onset voltage V_0, the conjunction of both field-dependent ionization and accumulation of mobile ions towards the interface can be producing a voltage-dependent mobility as

$$\mu = \mu_0 \frac{L_i}{L_D} \tag{S30}$$

where μ_0 is an effective mobility independent of field and position, L_i is Frenkel's equation[6] for the distance between the ions and their local potential maxima upon application of an external field

$$L_i = \sqrt{\frac{Q}{\epsilon_0 \epsilon_r \xi}} \tag{S31}$$

and L_D is the Debye length for the accumulation of mobile ions towards the electrodes

$$L_D = \sqrt{\frac{\epsilon_0 \epsilon_r k_B T}{Q^2 N_i}} \tag{S32}$$

In Equation (S32), N_i is the mobile ions concentration, k_B is the Boltzmann constant and T is the temperature. By substituting Equations (S31) and (S32) in (S30), and multiplying by $(L/L)^{1/2}$ we obtain

$$\mu = \mu_0 \sqrt{\frac{V_0}{L\xi}} \tag{S33}$$

where the onset voltage comes after

$$V_0 = \frac{Q^3 N_i L}{\epsilon_0{}^2 \epsilon_r{}^2 k_B T} \tag{S34}$$

The values for V_0 are presented in Figure S3 for a MAPI sample at room temperature. Note that one may expect values in the range 1-10 V for a 1.0 mm thickness sample, meaning that the concentration of mobile ions towards the interface is around 10^{14} cm^{-3}.

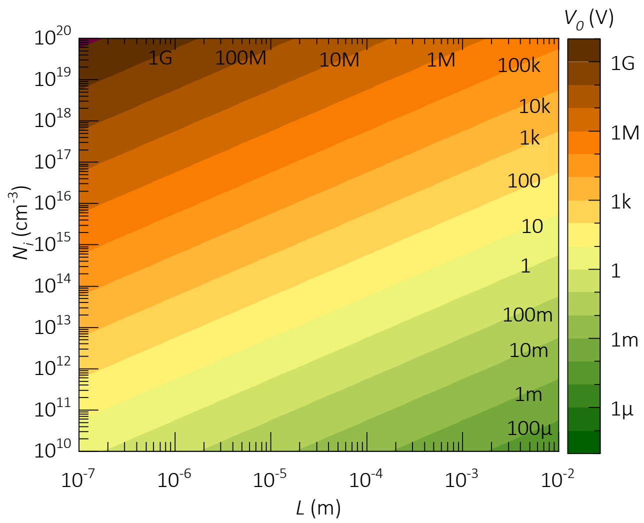

Figure S3: Onset voltage as a function of the distance between electrodes and the concentration of mobile ions towards the electrodes in the BVM regime of SCLC for a $MAPbI_3$ sample with ϵ_r=23,[3] $T = 300$ K and Q as the elementary charge in Equation (S34).

The BVM drift velocity can then be rewritten by substituting (S33) in (S29) as

$$v_d = \mu_0 \sqrt{\frac{V_0}{L}\xi} \tag{S35}$$

Alternatively, one can assume the BVM model as an approximation to a particular case of the Poole-Frenkel[6-8] ionized-trap-mediated transport when the field ξ and the charge carrier profile N meet certain specific criteria. For a start, we consider the Poole-Frenkel current density

$$J = \sigma_{PF} \exp\left[\frac{-q\phi}{k_B T}\right] \xi \exp\left[\sqrt{\frac{\xi}{\xi_{PF}}}\right] \tag{S36}$$

where σ_{PF} is the Poole-Frenkel conductivity, ϕ, the equilibrium potential barrier for the ionized traps and the Poole-Frenkel onset field is

$$\xi_{PF} = \frac{\pi \epsilon_0 \epsilon_r {k_B}^2 T^2}{Q^3} \tag{S37}$$

The electric field must fulfil two conditions for the BVM model to coincide with Equation (S36): *(i)* the field should be high enough that

$$\sqrt{\frac{\xi}{\xi_{PF}}} \gg 1 \tag{S38}$$

in order to decrease the potential barrier in the Poole-Frenkel effect, but *(ii)* only in a narrow field range where the current is not critically exponential and the Equation (S36) can be approximated to the McLaurin expression as

$$J \cong \sigma_1 \exp\left[\frac{-q\phi}{k_B T}\right] \xi \left(1 + \sqrt{\frac{\xi}{\xi_{PF}}}\right) \tag{S39}$$

where the effective BVM conductivity is taken as

$$\sigma_0 = \sigma_{PF} \exp\left[\frac{-q\phi}{k_B T}\right] \tag{S40}$$

In addition, the charge carrier distribution should be approximately constant as

$$N = N_0 \sqrt{\frac{L}{\xi_{PF} V_0}} \tag{S41}$$

where V_0 and L have the same meanings as in Equation (S34) and N_0 is a threshold effective conductivity for the transition from ohmic to the BVM regime of SCLC. Subsequently, assuming (S38) and substituting Equation (S41) in (S2) and (S39), the drift velocity can be approximated to Equation (S35) where the effective threshold mobility is assumed as

$$\mu_0 \cong \frac{\sigma_0}{N_0 Q} \tag{S42}$$

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