Electronic supplementary information

Unveiling the Synergistic Effect of A-Site Doping in Perovskite Nanosheets and Electrode Modulation for Boosting Dielectric Performance of Printed Microcapacitors

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Figure S1. AFM images of (a) $Sr_{1.9}Bi_{0.1}Nb_3O_{10}$ and (b) $Sr_{1.8}Bi_{0.2}Nb_3O_{10}$ nanosheet, respectively. Inset: line profiles across individual nanosheet.



Figure S2. The lateral flake size distribution images of (a) $Sr_2Nb_3O_{10}$, (b) $Sr_{1.9}Bi_{0.1}Nb_3O_{10}$, (c) $Sr_{1.8}Bi_{0.2}Nb_3O_{10}$, and (d) $Sr_{1.7}Bi_{0.3}Nb_3O_{10}$ nanosheets.

Table S1. The rheological parameters including viscosity, surface tension, density, and Z value for

| \mathbf{X}^{\prime} | |
|---|----|
| Viscosity (mPa·s) Surface tension (mN/m) Density (g/cm ³) Z values Z | le |
| SBNO 0.0 ink2.3126.50.8959.4 | |
| SBNO 0.1 ink2.3526.80.8959.3 | |
| SBNO 0.2 ink2.3726.20.8959.1 | |
| SBNO 0.3 ink2.3326.60.8959.3 | |
| Gr ink 11.3 33 0.94 2.2 | |
| Ag ink 7.5 25 1.0 2.98 | |

 $Sr_{2-x}Bi_xNb_3O_{10}$ (x = 0.0-0.3), graphene, and Ag ink.



Figure S3. (a) Surface and (b) cross-sectional SEM images of inkjet-printed $Sr_{1.7}Bi_{0.3}Nb_3O_{10}$ films on a quartz glass substrate.



Figure S4. The optical photographs of inkjet-printed (a) Ag/SBNO 0.0/Ag, (b) Ag/SBNO 0.3/Ag, (c) Gr/SBNO 0.0/Gr, and (d) Gr/SBNO 0.3/Gr microcapacitors on a glass substrate.



Figure. S5. SEM images of inkjet-printed Ag/SBNO 0.3/Ag and Gr/SBNO 0.3/Gr microcapacitors on a glass substrate.



Figure S6. Frequency dependence of (a, b) capacitance density, $\tan \delta$ and (c, d) impedance modulus values for inkjet-printed microcapacitors of (a, c) $Ag_B/Sr_{2-x}Bi_xNb_3O_{10}/Ag_T$ (x = 0.0-0.3) and (b, d) $Gr_B/Sr_{2-x}Bi_xNb_3O_{10}/Gr_T$ (x = 0.0-0.3), respectively.



Figure S7. Dielectric performance of inkjet-printed $Ag_B/Sr_{2-x}Bi_xNb_3O_{10}/Ag_T$ (x = 0.0-0.3) microcapacitors. Active area dependence of (a) capacitance and (b) capacitance density for inkjet-printed microcapacitors with a dielectric-layer thickness (t) of ~1.2 µm. (c) The curve of capacitance as a function of reverse thickness for inkjet-printed microcapacitors with the active area (s) of ~0.25 mm². (d) The curve of relative permittivity as a function of the ratio of active area to thickness.



Figure S8. Dielectric performance of inkjet-printed $Gr_B/Sr_{2-x}Bi_xNb_3O_{10}/Gr_T$ (x = 0.0-0.3) microcapacitors. Active area dependence of (a) capacitance and (b) capacitance density for inkjet-printed microcapacitors with a dielectric-layer thickness (t) of ~1.2 µm. (c) The curve of capacitance as a function of reverse thickness for inkjet-printed microcapacitors with the active area (s) of ~0.25 mm². (d) The curve of relative permittivity as a function of the ratio of active area to thickness.



Figure S9. AFM images of inkjet-printed (a) $Ag_B/Sr_2Nb_3O_{10}/Ag_T$, (b) $Ag_B/Sr_{1.9}Bi_{0.1}Nb_3O_{10}/Ag_T$, (c) $Ag_B/Sr_{1.8}Bi_{0.2}Nb_3O_{10}/Ag_T$, and (d) $Ag_B/Sr_{1.7}Bi_{0.3}Nb_3O_{10}/Ag_T$ microcapacitors on a glass substrate.



Figure S10. AFM images of inkjet-printed (a) $Gr_B/Sr_2Nb_3O_{10}/Gr_T$, (b) $Gr_B/Sr_{1.9}Bi_{0.1}Nb_3O_{10}/Gr_T$, (c) $Gr_B/Sr_{1.8}Bi_{0.2}Nb_3O_{10}/Gr_T$, and (d) $Gr_B/Sr_{1.7}Bi_{0.3}Nb_3O_{10}/Gr_T$ microcapacitors on a glass substrate.



Figure S11. Raman spectra of (a) $KSr_{2-x}Bi_xNb_3O_{10}$ and (b) $HSr_{2-x}Bi_xNb_3O_{10} \cdot nH_2O$ (*x* = 0, 0.1, 0.2, and

0.3) powders.



Figure S12. The surface resistivity of inkjet-printed $Sr_{2-x}Bi_xNb_3O_{10}$ (x = 0.0-0.3) film at different heat treatment temperatures.



Figure S13. (a-d) UV-vis absorption spectra and the corresponding Tauc curve of $Sr_{2-x}Bi_xNb_3O_{10}$ (x = 0.0-0.3) film printed on the quartz substrate. (e) XPS valence bands and (f) UPS spectra as the estimation of work functions for $Sr_2Nb_3O_{10}$ film printed on the quartz substrate.



Figure S14. Energy band diagrams at the (a) $Ag/Sr_2Nb_3O_{10}$ film and (b) graphene/ $Sr_2Nb_3O_{10}$ film interfaces, respectively.

Table S2. Lateral size statistics of $Sr_{2-x}Bi_xNb_3O_{10}$ (x = 0.0-0.3) perovskite nanosheets

| Nanosheets | $Sr_2Nb_3O_{10}$ | $Sr_{1.9}Bi_{0.1}Nb_3O_{10}$ | $Sr_{1.8}Bi_{0.2}Nb_3O_{10}$ | $Sr_{1.7}Bi_{0.3}Nb_3O_{10}$ |
|----------------------------|------------------|------------------------------|------------------------------|------------------------------|
| Percentage of lateral size | 78.3% | 70.7% | 72.3% | 76.7% |
| below 400 nm | | | | |



Figure S15. Electrical performance of inkjet-printed microcapacitors. Variation of capacitance as a function of bending cycles for inkjet-printed (a) $Ag_B/Sr_{2-x}Bi_xNb_3O_{10}/Ag_T$ (x = 0.0-0.3) and (c) $Gr_B/Sr_{2-x}Bi_xNb_3O_{10}/Gr_T$ (x = 0.0-0.3) microcapacitors, respectively. Variation of capacitance as a function of temperature for inkjet-printed (b) $Ag_B/Sr_{2-x}Bi_xNb_3O_{10}/Ag_T$ (x = 0.0-0.3) and (d) $Gr_B/Sr_{2-x}Bi_xNb_3O_{10}/Ag_T$ (x = 0.0-0.3) and (d) $Gr_B/Sr_{2-x}Bi_xNb_3O_{10}/Gr_T$ (x = 0.0-0.3) microcapacitors, respectively.



Figure S16. The optical photographs of $Ag_B/Sr_{1.7}Bi_{0.3}Nb_3O_{10}/Ag_T$ microcapacitors treated in (a) air, (b) water, (c) ethanol, (d) acetonitrile, (e) NMP, (f) DMF, (g) 1,2-dichlorobenzene, and (h) n-hexane.



Figure S17. The optical photographs of $Gr_B/Sr_{1.7}Bi_{0.3}Nb_3O_{10}/Gr_T$ microcapacitors treated in (a) air, (b) water, (c) ethanol, (d) acetonitrile, (e) NMP, (f) DMF, (g) 1,2-dichlorobenzene, and (h) n-hexane.



Figure S18. Electric field dependence of the capacitance variation for inkjet-printed $Ag_B/Sr_{2-x}Bi_xNb_3O_{10}/Ag_T$ (x = 0.0, 0.2) and $Gr_B/Sr_{2-x}Bi_xNb_3O_{10}/Gr_T$ (x = 0.0, 0.2) microcapacitors.

| Solvent | Polarity | Polarity index | Dielectric constant | Molecular weight (g/mol) |
|--------------|---------------|----------------|---------------------|--------------------------|
| Water | Polar Protic | 10.2 | 80.1 | 18.01524 |
| Ethanol | Polar Protic | 4.3 | 25.3 | 46.06844 |
| Acetonitrile | Polar Aprotic | 6.2 | 36.64 | 41.052 |
| NMP | Polar Aprotic | 6.7 | 32.55 | 99.13 |
| DMF | Polar Aprotic | 6.4 | 38.25 | 73.095 |
| DCB | Non polar | 2.7 | 9.32 | 147.01 |
| Hexane | Non polar | 0.06 | 1.89 | 86.175 |

Table S3. Physical and chemical properties of different types of solvents¹⁻³.

Table S4. The yield and reproducibility of inkjet-printed microcapacitors.

| Sample quantity | Working [Vorking] | Yield and Reproducibility |
|-----------------|---|--|
| <mark>15</mark> | <mark>12</mark> | <mark>80%</mark> |
| <mark>15</mark> | <mark>14</mark> | <mark>93.3%</mark> |
| <mark>15</mark> | <mark>13</mark> | <mark>86.7%</mark> |
| <mark>15</mark> | <mark>13</mark> | <mark>86.7%</mark> |
| <mark>15</mark> | <mark>9</mark> | <mark>60%</mark> |
| <mark>15</mark> | <mark>10</mark> | <mark>66.7%</mark> |
| <mark>15</mark> | <mark>11</mark> | <mark>73.3%</mark> |
| <mark>15</mark> | <mark>10</mark> | <mark>66.7%</mark> |
| 120 | <mark>92</mark> | <mark>76.7%</mark> |
| | Sample quantity 15 15 15 15 15 15 15 15 15 15 | Sample quantity Working 15 12 15 14 15 13 15 13 15 9 15 10 15 11 15 10 15 10 15 10 15 92 |

References:

- 1. C. M. Hansen, *Hansen solubility parameters: a user's handbook*, CRC Press, 2007.
- 2. D. R. Lide, *CRC handbook of chemistry and physics*, CRC Press, 2004.
- 3. C. L. Yaws, *Thermophysical properties of chemicals and hydrocarbons*, William Andrew, 2008.