# Efficient External Electric Field Manipulated Nonlinear Optical Switches of All-Metal Electride Molecules with Infrared Transparency: Nonbonding Electron Transfer Forms an Excess Electron Lone Pair

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**Table S1** First hyperpolarizability ($\beta_0^e(F)/10^3$au) at MP2/6-311+g(3df)/LANL2DZ level, the transition energy $\Delta E$ (eV), the oscillator strength $f_0$, and the difference in the dipole moment between the ground and the crucial Excited State $\Delta\mu$ (D) from the CIS/6-311+g(2d)/LANL2DZ level for all-metal electride molecule Be(Ni@$Pb_{12}$)Be.

| F/$10^{-4}$au | $f_0$ | $\Delta E$ | $\Delta\mu$ | $\beta_0$ ($10^3$au) |
|---|---|---|---|---|
| 0 | 0.1995 | 1.4536 | 0.049 | 0 |
| 5 | 0.1906 | 1.4597 | 1.376 | 4 |
| 10 | 0.1647 | 1.4842 | 2.704 | 10 |
| 15 | 0.1419 | 1.5184 | 3.433 | 14 |
| 20 | 0.1235 | 1.5596 | 3.917 | 17 |
| 25 | 0.3223 | 2.9609 | 6.406 | 19 |
| 30 | 0.3636 | 2.9203 | 8.895 | 20 |
| 35 | 0.3631 | 2.8727 | 10.485 | 22 |
| 40 | 0.3651 | 2.8105 | 9.791 | 24 |

**Table S2** First hyperpolarizability ($\beta_0^e(F)/10^3$au) at MP2/6-311+g(3df)/LANL2DZ level, the transition energy $\Delta E$ (eV), the oscillator strength $f_0$, and the difference in the dipole moment between the ground and the crucial Excited State $\Delta\mu$ (D) from the CIS/6-311+g(2d)/LANL2DZ level for all-metal electride molecule Mg(Ni@$Pb_{12}$)Mg.

| F/$10^{-4}$au | $f_0$ | $\Delta E$ | $\Delta\mu$ | $\beta_0$ ($10^3$au) |
|---|---|---|---|---|
| 0 | 0.4842 | 1.2353 | 0 | 0 |
| 5 | 0.4072 | 1.2681 | 1.947 | 110 |
| 10 | 0.3555 | 1.3002 | 2.627 | 50 |
| 15 | 0.5081 | 3.1048 | 13.077 | 16 |
| 20 | 0.5236 | 3.0498 | 2.422 | 2 |
| 25 | 0.4869 | 2.9843 | 16.089 | 9 |
| 30 | 0.3843 | 2.9204 | 13.333 | 9 |
| 35 | 0.2900 | 2.6901 | 2.360 | 7 |
| 40 | 0.1126 | 2.3394 | 0.044 | 1 |

**Table S3** First hyperpolarizability ($\beta_0^e(F)/10^3$au) at MP2/6-311+g(3df)/LANL2DZ level, the transition energy $\Delta E$ (eV), the oscillator strength $f_0$, and the difference in the dipole moment between the ground and the crucial excited etate $\Delta\mu$ (D) from the CIS/6-311+g(2d)/LANL2DZ level for all-metal electride molecule Ca(Ni@$Pb_{12}$)Ca.

| F/$10^{-4}$au | $f_0$ | $\Delta E$ | $\Delta\mu$ | $\beta_0$ ($10^3$au) |
|---|---|---|---|---|
| 0 | 1.7903 | 1.1653 | 0 | 0 |
| 5 | 1.7057 | 1.1560 | 1.934 | 1700 |
| 10 | 1.4975 | 1.1367 | 2.325 | 2100 |
| 15 | 1.2543 | 1.1260 | 0.615 | 1300 |
| 20 | 1.0447 | 1.1404 | 2.422 | 220 |
| 25 | 0.8982 | 1.1829 | 5.201 | 320 |
| 30 | 0.8171 | 1.2420 | 5.876 | 420 |
| 35 | 0.8673 | 2.7506 | 3.849 | 340 |
| 40 | 0.8331 | 2.7393 | 2.970 | 200 |

**Table S4** Effects of external electric field ($F/10^{-4}$au) on the static first hyperpolarizability $\beta_0^e(F)/10^3$au) for the optimized structures under $F$.

| $F/10^{-4}$au | Be(Ni@$Pb_{12}$)Be | Mg(Ni@$Pb_{12}$)Mg | Ca(Ni@$Pb_{12}$)Ca |
|---|---|---|---|
| 0 | 0 | 0 | 0 |
| 2 | 1 | 15 | 700 |
| 4 | 3 | 118 | 1500 |
| 5 | 4 | 106 | 1700 |
| 6 | 5 | 93 | 1900 |
| 8 | 8 | 70 | 2200 |
| 10 | 10 | 50 | 2100 |
| 12 | 11 | 35 | 1900 |
| 14 | 13 | 21 | 1500 |
| 15 | 14 | 16 | 1300 |
| 16 | 14 | 12 | 1000 |
| 18 | 16 | 4 | 600 |
| 20 | 17 | 2 | 220 |
| 25 | 19 | 9 | 320 |
| 30 | 20 | 9 | 420 |
| 35 | 22 | 7 | 340 |
| 40 | 24 | 1 | 200 |