# Supplemental Material: Electronic correlation in nearly free electron metals with beyond-DFT methods

Subhasish Mandal,[1,2] Kristjan Haule,[2] Karin M. Rabe,[2] and David Vanderbilt[2]

[1] *Department of Physics and Astronomy, West Virginia University, Morgantown, WV, USA*
[2] *Department of Physics and Astronomy, Rutgers University, Piscataway, NJ, USA*

## SUPPLEMENTARY METHODS

**Crystal Structures:** The experimental crystal structures are obtained from the ICSD-database. The ICSD numbers are given in Supplementary Table I.

**GW-details:** The number of unoccupied bands and the number of core states included in the GW calculations are described in Supplementary Table II

## SUPPLEMENTARY DISCUSSION

First, in Supplementary Figure 1, we show the Coulomb $U$ dependence of eDMFT spectral function for elemental Na. The bandwidth shows a strong dependence on the Coulomb $U$. Pink dots in Supplementary Figure 1 indicate angle-resolved-photoemission (ARPES) data from Lyo and Plummer, Phys. Rev. Lett. 60, 1558 (1988).

The imaginary part of the eDMFT self-energy gives the scattering rate of electrons and also gives additional microscopic insights into the correlated electronic structure evolution. In Supplementary Figure 2, we show the frequency dependence of the imaginary part of the self-energy for s-orbital in Mg, Ca, Sr, and Rb.

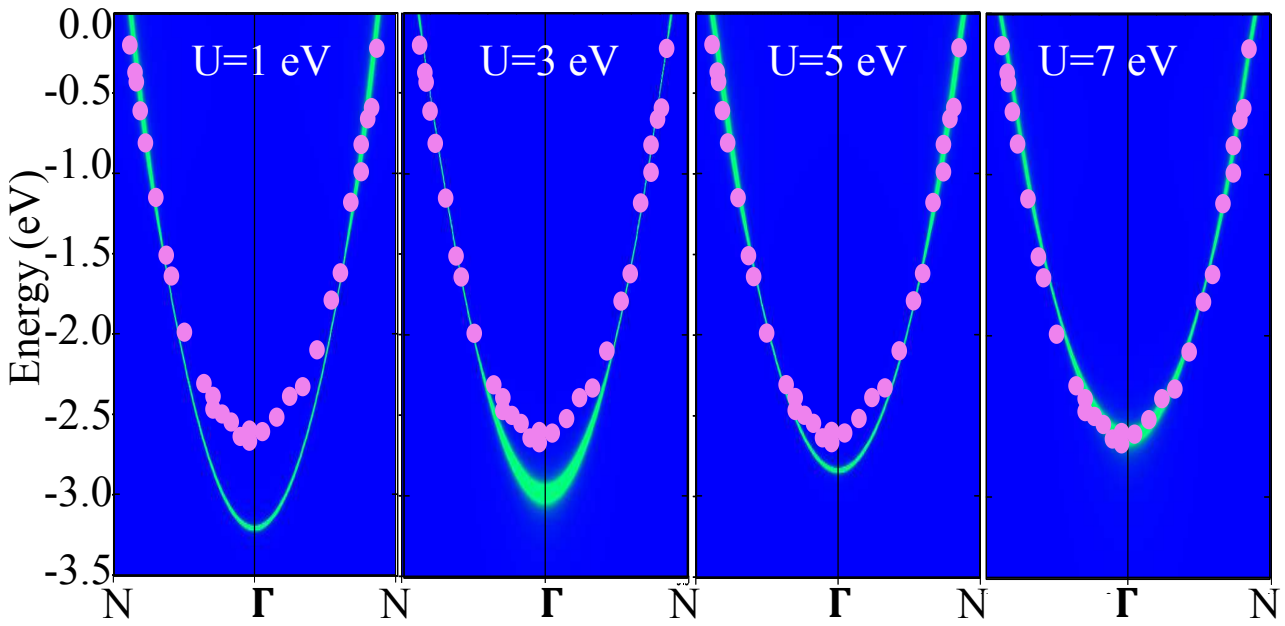
In Supplementary Figures 3-7, we describe full band structures for elemental Na, K, Mg Be, Sr, Cs, Ca, and Rb respectively as computed in LDA, mBJ, $G_0W_0$, B3LYP, and eDMFT in various energy windows. They clearly show the differences in bandwidths for occupied as well as unoccupied bands. The renormalization of the bands also shows a strong K-dependence as the differences in bandwidth among various methods vary strongly in various parts of the Brillouin zone. The difference is always largest at the high-symmetric Γ-point. We find B3LYP not only overestimates bandwidths of occupied bands, but it also does the same for the unoccupied bands. Again, LDA and mBJ bands disperse very similarly. For K, Sr, Cs we notice the bandwidths are reduced in GW compared to LDA for occupied bands but unoccupied bands, the bandwidths in GW are larger than that of LDA or mBJ.

| Compound | ICSD-ID |
|---|---|
| Li | 44367 |
| Be | 1425 |
| Na | 196972 |
| Mg | 76748 |
| K | 44670 |
| Ca | 44348 |
| Rb | 44869 |
| Sr | 76162 |
| Cs | 42662 |
| Ba | 96587 |

Supplementary Table I. ICSD-IDs for compounds studied here.

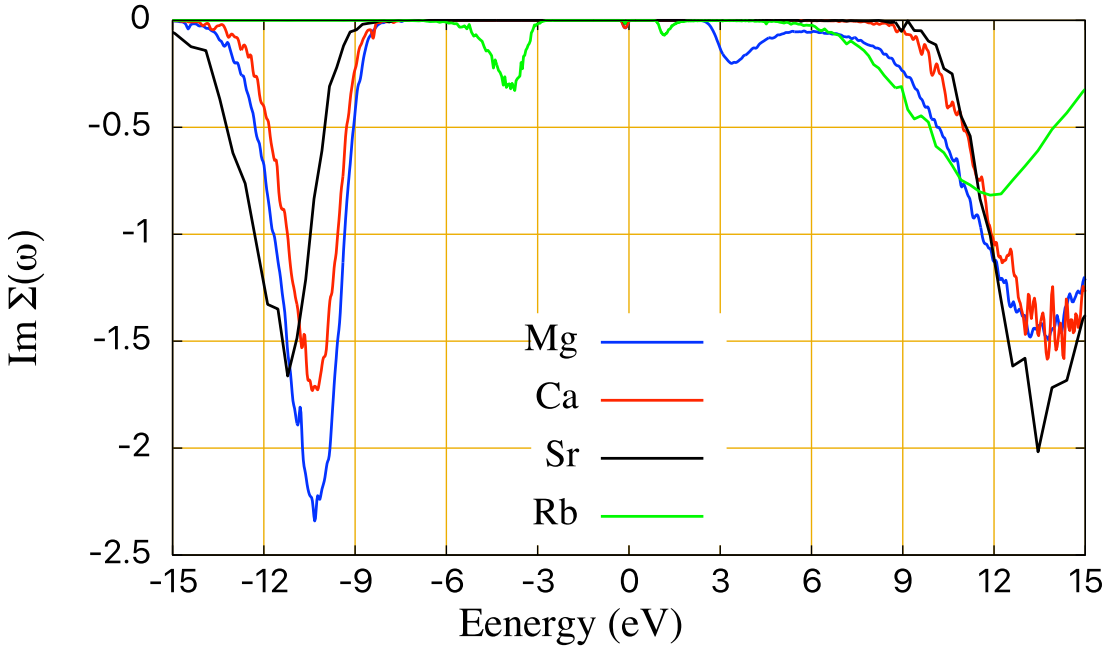
| Compound | #unoccupied bands | #core state |
|---|---|---|
| Li | 40 | 0 |
| Be | 62 | 2 |
| Na | 88 | 1 |
| Mg | 126 | 2 |
| K | 188 | 8 |
| Ca | 109 | 8 |
| Rb | 225 | 25 |
| Sr | 143 | 25 |
| Cs | 177 | 32 |
| Ba | 225 | 25 |

Supplementary Table II. Number of unoccupied bands and core states considered for constructing Polarizability matrix in GW calculations.

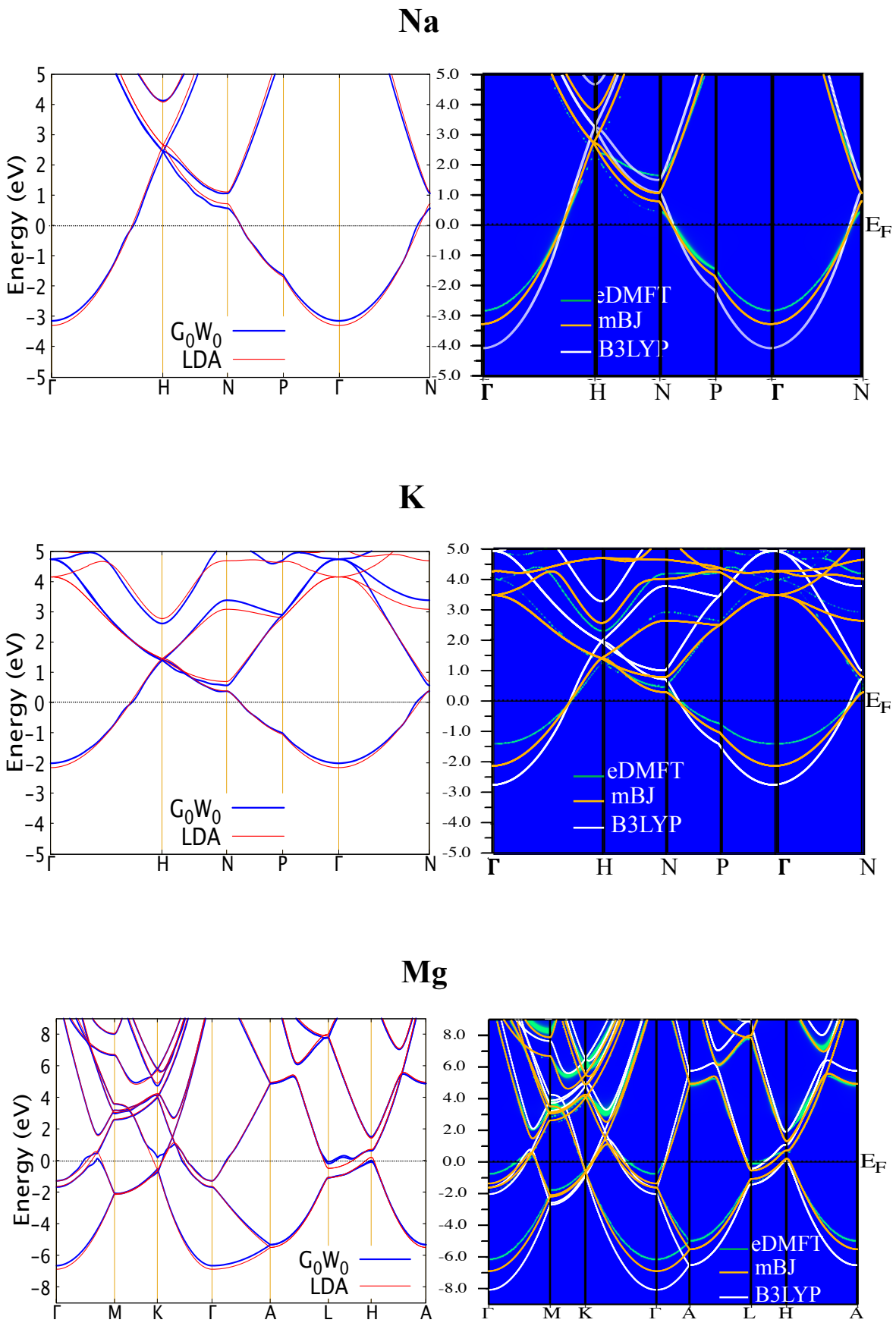



Supplementary Figure 1. The dependence of Coulomb $U$ in eDMFT spectral function for elemental Na. Pink dots indicate angle-resolved-photoemission (ARPES) data from Lyo and Plummer, Phys. Rev. Lett. 60, 1558 (1988).

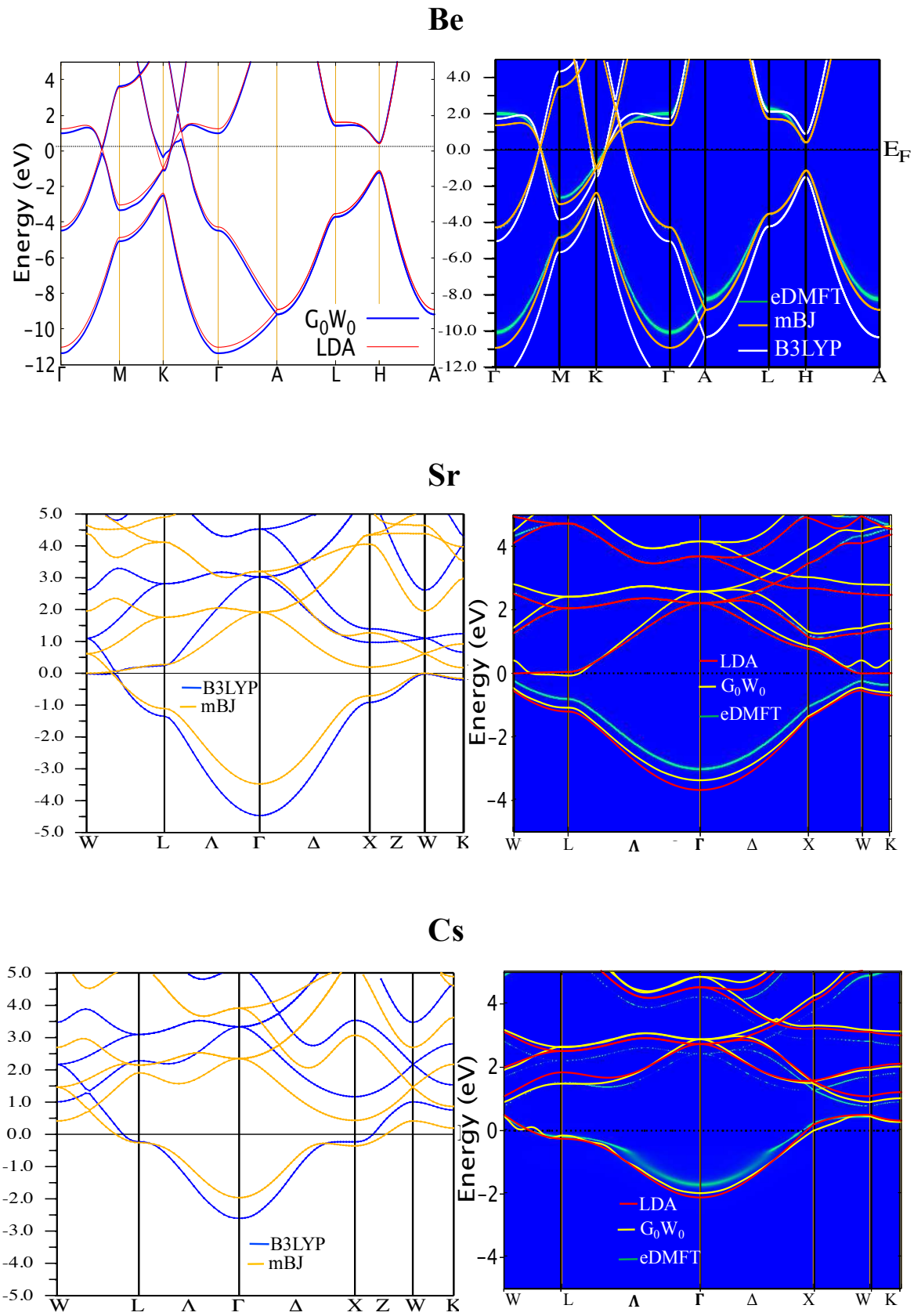
## SUPPLEMENTARY FIGURES



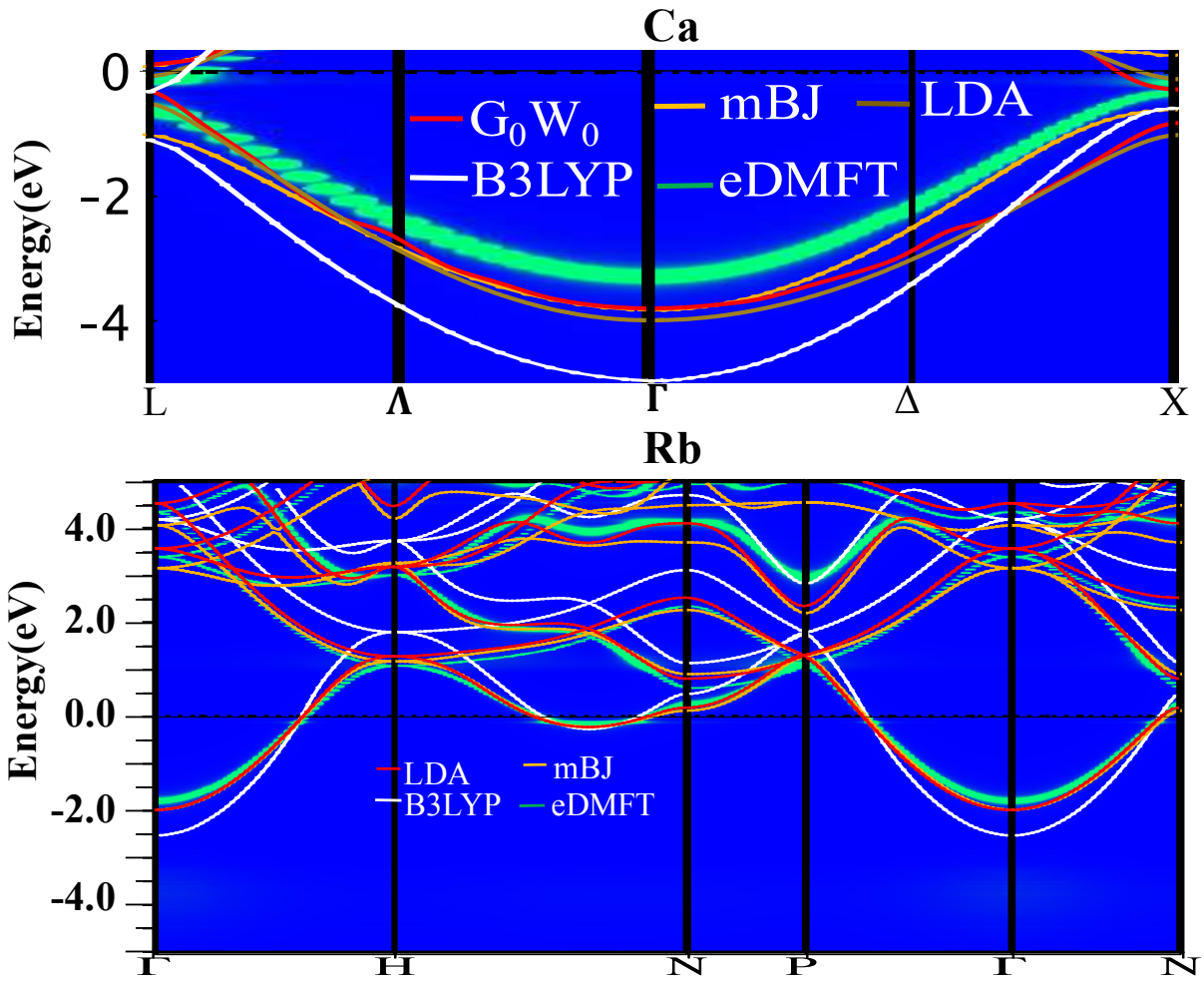

Supplementary Figure 2. Imaginary part of the eDMFT self-energy of Mg, Ca, Sr, and Rb




Supplementary Figure 3. Full band structures of elemental Na (top), K (middle) and Mg (bottom) as computed in LDA, mBJ, $G_0W_0$, B3LYP, and eDMFT.




Supplementary Figure 4. Band structures of elemental Be (top), Sr (middle) and Cs (bottom) as computed in LDA, mBJ, $G_0W_0$, B3LYP, and eDMFT.




Supplementary Figure 5. Band structures of elemental Ca (top) and Rb (bottom) as computed in LDA, mBJ, $G_0W_0$, B3LYP, and eDMFT.