II. COMPUPATIONAL DETAILS

Full-electron density functional theory (DFT) implemented in the Dmol^3 package [17, 18] is used in our calculations. The Perdew, Burke, and Ernzerhof (PBE) [19] functional is used for exchange and correlation interaction in the generalized gradient approximation(GGA). The basis set is composed of double numerical basis and a polarized function (DNP) is chosen. The supercell is chosen as $20\times20\times c$ Å^3, where c is the lattaice constant along the periodic direction and is varied for different TM cases [see Table I]. The corresponding Brillouin zone is sampled by a $1\times1\times50$ Monkhorst and Pack grid.[20] All structures are fully relaxed.

In order to investigate the different magnetic configurations and Peierls transition in $[TM_2(Ant)]_\infty$, a supercell containing four TMs and two Ants are used. For each kind of $[TM_2(Ant)]_\infty$, as shown in Figs. 1(b)-(f), five magnetic configurations are considered. The combination of two characters XY (X and Y correspond to N(non-magnetic), F(ferromagnetic) and A(anti-ferromagnetic)) are adopted for representing the related magnetic configurations, with X and Y describing the intralayer and interlayer magnetic coupling of TM atoms, respectively. For example, as shown in Fig. 1(d), *FA* means the ferromagnetic coupling between the inner layer TM atoms and the anti-ferromagnetic coupling between two nearest neighbor layers.

III. RESULTS AND DISCUSSIONS

After full relaxation, the calculated structural, electronic and magnetic properties of $[TM_2(Ant)]_\infty$ are summarized in Table I. Peierls transition does not occur in $[TM_2(Ant)]_\infty$, and no magnetic moment is observed in the cases of $[Sc_2(Ant)]_\infty$ and $[Mn_2(Ant)]_\infty$. In contrast, Ti and Fe are spin polarized, but the magnetic ground states of $[Ti_2(Ant)]_\infty$ and $[Fe_2(Ant)]_\infty$ are *FA* and *AF*, respectively. Hence, the total magnetic moments of $[Ti_2(Ant)]_\infty$ and $[Fe_2(Ant)]_\infty$ supercells are zero. In addition, our calculations predict that *FF* is the ground state of both $[V_2(Ant)]_\infty$ and $[Cr_2(Ant)]_\infty$, and that the supercells of $[V_2(Ant)]_\infty$ and $[Cr_2(Ant)]_\infty$ respectively possess a magnetic moment of $2.00\mu_B$ and $0.96\mu_B$ per TM ion, with a finite negative magnetic moment formed on the Ant part. Further analysis shows that $[V_2(Ant)]_\infty$ and $[Cr_2(Ant)]_\infty$ are half-metallic ferromagnets which can be seen from