Electronic Supplementary Information

The electronic and magnetic properties of transition-metal elements doped three-dimensional topological Dirac semimetal in Cd_3As_2

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Determine the effective onsite Coulomb interaction parameter (*U*)

In this work, we have performed calculations with *U* ranging from 2 eV to 6 eV. The results, as the example of Cr shown in **Fig. S1**, indicate that *U* value has little influence on the half-metallic properties of Cr-doped Cd_3As_2. While the magnetic moment of Cr is slightly modified, from 3.75 μ_B for U= 2eV, to 4.0 μ_B for U=4 eV and 4.25 μ_B for U=6 eV. As discussed in the paper, it is expected that Cr shows 4 μ_B in our system. Therefore, U=4 eV could be the best choice for Cr. Moreover, the TM atoms are tetrahedrally coordinated in our studied system, which are very similar to the cases in III−V and II−VI diluted magnetic semiconductors. In these compounds, the estimated *U* values for 3*d* orbitals according to the photoemission spectra vary between 3.5 and 4.5 eV depending on the used theoretical models.[1-4] Therefore, to keep consistence of

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our results, it is reasonable to take a value of U = 4.0 eV for all investigated TMs in this study.

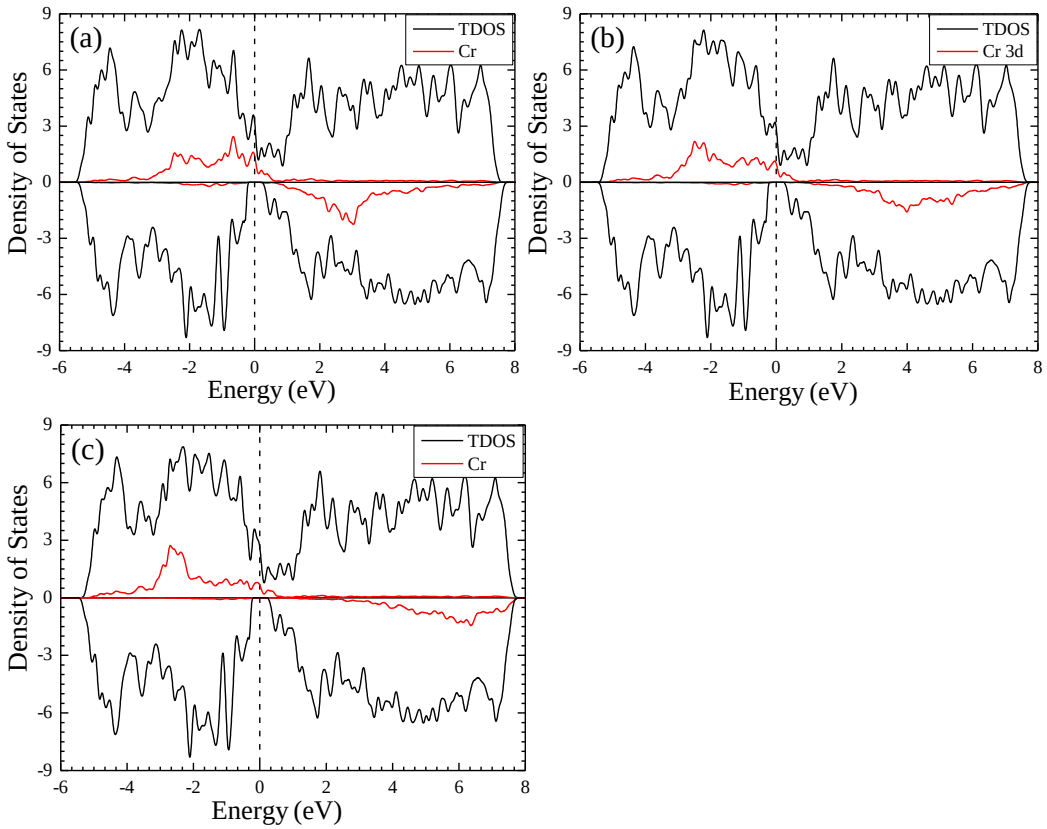

Fig. S1. DOS for Cr-doped Cd_3As_2 with (a) U=2 eV, (b) U=4 eV, and (c) U=6 eV.

Monte Carlo Simulations of Ising Model:

1. Ising Model

The Ising Model is a simple model of a solid that exhibits a phase transition resembling ferromagnetism. In this model, each spin direction may be either "spin-up" (+1) or "spin-down" (-1). though generalized models may include long-range interactions and more choices for spin direction. The Ising Hamiltonian of the system can be described as:

$$H = -\sum_{i,j} J_{ij} m_i m_j$$

where J_{ij} is is the exchange coupling constant between the i^{th} and j^{th} Cr atoms, taken from the DFT calculations as shown in **Figure 6a**.

2. The Algorithm of Monte Carlo

The Monte Carlo simulations are carried out using a $10 \times 10 \times 10$ supercell with periodic boundary conditions. The algorithm is described as follows:

1. A trial configuration is made by randomly choosing one spin;

2. The energy difference of the trial state relative to the present state, ΔE, is calculated;

3. If $\Delta E \leq 0$, the trial state is energetically favorable and thus accepted. Otherwise, a random number $0 \leq \delta \leq 1$ is generated, and the new state is only accepted if $\exp(-\beta \Delta E) > \delta$, where $\beta = 1/k_B T$;

The calculations last for 8×10^6 loops. The system is allowed to equilibrate for a long time (2×10^6 loops in this study). The magnetization is then measured by taking the sum of all the spins in the lattice. For high temperatures, the spins remain randomly aligned after long periods of equilibration, whereas for low temperatures, the spins end up pointing in mostly the same direction. The Curie temperature (T_c) is determined at the transition point.

Reference:

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