**First-principles scheme for strongly correlated electron systems with maximally localized Wannier functions: Application to black sodalite**

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In *ab initio* calculation for strongly correlated materials which combines model calculation and density functional calculation, constructing an effective model from realistic electronic structure calculation is one of the most important steps. Recently, a variety of methods for this "downfolding" procedure have been proposed, and have been applied to various correlated materials such as transition metals, transition metal oxides etc.

On the other hand, reliable downfolding has not been performed for materials for which the localized basis of the effective model extends beyond a single site in a non-trivial manner. For example, although various interesting many-body phenomena observed in organic compounds such as BEDT-TTF salts have been studied in terms of Hubbard-like models, *ab inito* estimation of interaction parameters like Hubbard $U$ is yet to be performed. To overcome this problem, recently, two of the present authors and their collaborators have developed a new method to estimate $U$ by using maximally localized Wannier functions (MLWFs) [1].

In order to demonstrate how this new method works well for materials which have been problematic so far, here we apply it to black sodalite, $Na_8(AlSiO_4)_6$. For this material, it is well known that a periodic array of $Na_4^{3+}$ clusters loaded in the aluminosilicate cages forms narrow bands in the wide band gap, and the system can be mapped to the half-filling Hubbard model or the S=1/2 Heisenberg model (Fig.1) [2].This material is interesting in that it is magnetic while only non-magnetic elements comprised of this material.

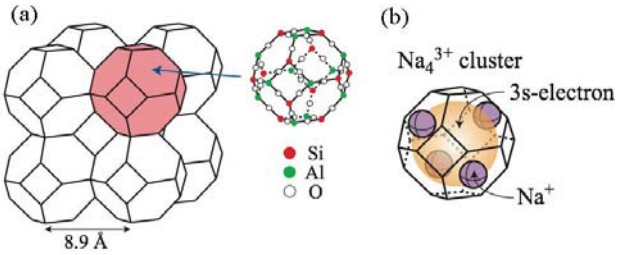


Fig. 1: Crystal structure of black sodalite. The frame is comprised of Si, Al, O (a), while the main contribution to the energy bands around the Fermi level comes from $Na_4^{3+}$ clusters in the aluminosilicate cages (b).

While the bare interaction parameter was estimated in Ref. [2], the (screened) Hubbard $U$ for the $Na_4^{3+}$ cluster has not been evaluated. In this study, we construct the MLWF for the $Na_4^{3+}$ cluster and estimate $U$ for it. Then we derive an effective spin model $H = \Sigma_{ij} J_{ij} S_i S_j$, for which we find $J$=28.2K for the nearest neighbours and $J$=8.6K for the next-nearest neighbours.

The Neel temperature ($T_N$) and the Weiss temperature ($\theta$) calculated by the mean field approximation are 87K and 139K, which are in good agreement with experimental values, $T_N$ =55K and $\theta$ =170K. We also calculate the uniform spin susceptibility as a function of the temperature by the Pade approximation of the high temperature expansion up to the 8th order (Fig.2). We can see that the theoretical and experimental result show an excellent agreement, suggesting that our downfolding method indeed works successfully.

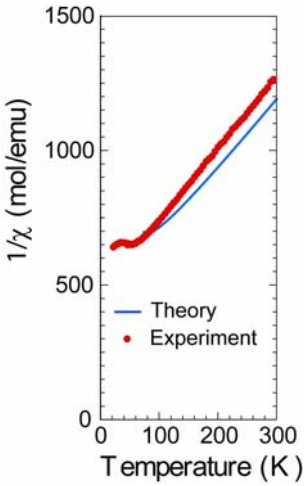


Fig.2: Uniform spin susceptibility calculated by the Pade approximation of the high temperature expansion (blue line) compared with the experimental results (red circles).

[1] K. Nakamura *et al*, Phys. Rev. B **74** 235113 (2006).
[2] O.F. Sankey *et al*, Phys. Rev. B **57** 15129 (1998).