First-principles calculations of thermoelectric properties in magnetic materials

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1. *Development of Methods and Codes*

The thermoelectric conversion based on the anomalous Nernst effect (ANE) has attracted much attention because the ANE realizes the high-density integration more easily compared to that based on the Seebeck effect[1]. The ANE is the phenomenon that the electric power is created along the direction perpendicular to both the temperature gradient and the magnetization in magnetic materials. This effect is induced by the anomalous Hall conductivity (AHC), and if the AHC changes drastically as a function of the Fermi level, we expect the large ANE[2]. In our previous first-principles study, we predicted that an electron doped EuO skyrmion crystal shows the large ANE induced by the finite Chern number at the Fermi level[3]. For discovery and design of new materials which have the large ANE via computational high throughput screening, we need an efficient computational method to investigate the Fermi level dependence of the AHC. We have implemented the code of computing the AHC applicable to metallic systems in OpenMX package[4] by improving Fukui-Hatsugai-Suzuki method[5]. Using this code, we computed AHCs $\sigma_{xy}(\varepsilon)$ and transverse thermoelectric conductivities $\alpha_{xy}(\varepsilon)$ in two-dimensional ferromagnetic materials.

2. *Applications to 2D Materials*

The discovery of magnetism in the two-dimensional materials[6] open a way for realizing thermoelectric devices based on anomalous Nernst effect. The surface phonon scattering advantage in the two-dimensional materials can decrease the thermal conductivity which can increase the thermoelectric figure of merit ZT[7].The magnitude of anomalous Nernst effect itself depend on Seebeck effect or anomalous Hall effect[2, 3]. In our previous study, we reported that the half-metallic materials possess large anomalous Nernst effect[8]. Here, we performed first-principles calculations to explore the anomalous Nernst effect of half-metallic transition-metal dihalides monolayer using OpenMX package[4]. We analyzed its magnitude based on the semiclassical transport theory and found that there is large anomalous Nernst effect at 100 K. The large Berry curvature which contributed by bands near Fermi level is the main reason of its large magnitude. These results suggest that transition-metal dihalides monolayer can potentially be used in thermoelectric devices.

3. *Thermal Conductivity and ZT*

To evaluate efficiency of thermoelectric materials, it is important to examine the contribution of thermal conductivity to the thermoelectric figure of merit ZT that represents the performance of the material. Since the thermal conductivity can be described for each electron and lattice, in this research, we theoretically computed both electronic and lattice thermal conductivity, and then evaluated ZT. Theoretical analysis of lattice thermal conductivity is conducted by Boltzmann transport the-

ory with relaxation time approximation. In order to do this, it is necessary to determine the inharmonic atomic force constant exactly, and an accurate analysis is carried out by first-principles calculations based on density functional theory. As computational codes, OpenMX package[4] is used as a density functional calculation, and ALAMODE[9, 10] is used as a lattice thermal conductivity calculation. The half-Heusler compounds are ternary compounds, have composition formula of XYZ, and are expected as thermoelectric materials that exhibit high power factor in a medium temperature range of about 600 to 1000 K. We analyzed thermoelectricity and thermal conductivity of ferromagnetic half-metallic half-Heusler MnCoSb[8]. We evaluated the ZT for the Seebeck and anomalous Nernst effect. We discussed carrier and temperature dependence of ZT.

4. *Surface Structures and Atomic Radii*

To design new functional materials, surface alloying is an effective way to obtain desired materials. Surface alloys are synthesized by inserting atoms into the surface. This topic has attracted a lot of research attention lately because of some of its applications, such as spintronics[11], where there is a giant spin splitting in Bi/Ag(111) surface alloys[12, 13]. Many types of surface alloys have been studied: for example, Ge/Ag(111), Pb/Ag(111), Bi/Ag(111), and Sn/Ag(111). One structure that has been observed in experiments with surfaces is $\sqrt{3} \times \sqrt{3}$ R 30°. Structural studies for surfaces alloys can provide important information for forming metal-semiconductors interface and metal interfaces. The metal-semiconductor interface is very important to industry, for example, the Si/Ag interface is a promising candidate for solar cells. We systematically explored the structural stability of M/Ag(111) surface alloys by first-principles calculation using the OpenMX package[4], where M is group III (B, Al, Ga, In and Tl), IV (C, Si, Ge, Sn and Pb) and V atom (N, P, As, Sb and Bi). We focused on the corrugation parameter d which is determined by the height of the M atom from the Ag atom in the plane of the top-most atom. We introduced concept of atomic radii to understand the tendencies of corrugation in M/Ag(111). The formation energy for each M atom is also calculated to determine the stability of the surface alloys. We discussed the dependence of M atoms on corrugation parameters.

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