Development of Fast Calculation Codes of Berry Phases in First-principles Calculations and its Application to Energy Conversion Materials

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This year further development of calculation methods and programs for intrinsic anomalous Hall conductivity (AHC) was carried out. AHC is related to thermoelectric conversion phenomena such as the anomalous Nernst effect, and its intrinsic contribution is based on the Berry curvature of the electronic states of the material. Since the previous year, we have also been developing a computational method for analyzing the layer-by-layer contribution of the AHC by combining the local Berry phase (LBP) method with the hybrid Wannier function method. We have performed applied calculations on various systems such as magnetic topological insulators and magnetic metal thin films and confirmed that the layer-by-layer AHC contribution in magnetic topological insulators and magnetic metal thin films can be analyzed.

On the other hand, in complex material systems, especially those that have defects and impurities, many degenerate points occur due to folding of the electronic band structure in the first Brillouin zone (FBZ), and obtaining the intrinsic AHC is computationally difficult because it requires more k-point sampling than that for perfect crystals. We have developed the LBP method [1] by extending the Fukui-Hatsugai-Suzuki method [2] to metallic systems for intrinsic AHC calculations. In the LBP method, the calculation of the determinant of the overlap matrix that consists of the periodic parts of the Bloch functions between k-points on the edges of the plaquettes is necessary, so basically the occupation numbers within the plaquettes must be the same. When the chemical potential crosses bands, the occupation numbers differ near the intersection point, and they can be approximated by averaging. If there are degenerate points within the plaquette, coarse k-point sampling may result in only one of the bands at a degenerate point contributing to the determinant, leading to numerical instability.

Then, we implemented various methods for calculating determinants in the LBP method using the first-principles calculation code OpenMX [3]. We found that even when degenerate points exist due to the folding of the FBZ, it is possible to evaluate appropriately by successively assessing the properties of overlapping matrices and then providing appropriate minor determinants. We have shown that the proposed method is effective by performing AHC calculations for BCC iron and its supercell systems. In the conventional LBP method, the AHC vibration increased due to the superlattice formation of cells when sampling was performed with the almost same k-point spacing. In the above new method, in BCC iron, it was found that the degree of fluctuation in the results obtained using the primitive cell model, conventional cell model, and supercellized 4-atom model was equivalent, indicating that the AHC vibration, which appears as an error due to superlattice formation of cells, was suppressed. Excessive k-point sampling is no longer necessary, and the required number of k-points can be easily estimated, so it is expected that high-throughput calculations will also be possible. We have developed a new LBP method and have confirmed that the numerical instability can be resolved for various superlattice systems.

References

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