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Theory of Orbital Magnetic Quadrupole Moment and Magnetoelectric Susceptibility

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In classical electromagnetism, electric and magnetic multipole moments characterize anisotropy of the charge and charge current densities. When electric or magnetic dipole moments align in a certain direction, the system is called a ferroelectrics or ferromagnet. Higher-order multipole moments have been studied traditionally in strongly correlated electron systems and more recently in higher-order topological insulators. Thus, multipole moments are more ubiquitous than in the nineteenth century when classical electromagnetism was established.

Among multipole moments, the magnetic quadrupole moment (MQM) has been believed to be an important ingredient for the magnetoelectric (ME) effect. However, there is no microscopic or quantitative discussion in the literature because multipole moments involve the unbounded position operator and seem ill-defined in periodic crystals.

Here we derive a quantum-mechanical formula of the orbital MQM in periodic crystals [1]. First, we define MQM and prove a direct relation to the ME susceptibility based on thermodynamic relations. This relation indicates that MQM is a microscopic origin of the ME effect. Second, we calculate the orbital MQM in the Bloch basis using the gauge-covariant gradient expansion of the Keldysh Green's function. Finally, we apply these results to antiferromagnetic semiconductors BaMn2As2 and CeMn2Ge2. We find that the orbital contribution to the ME susceptibility is comparable with or even dominant over the spin contribution.

[1] Atsuo Shitade, Hikaru Watanabe, and Youichi Yanase, arXiv: 1803.00217.

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