

Microscopic Formulation and Group-Theoretical Analysis of Circular Raman Optical Activity in Spin Systems

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Raman scattering has been widely used to probe excitations in materials. Recently, theoretical studies have suggested that circular Raman optical activity (CROA), the scattering-intensity difference between left- and right-circularly polarized light, can probe the quantum geometry of magnon bands [1,2] and of spin liquid systems [3]. Moreover, CROA might be used to distinguish between magnetic domains.

Although symmetry analysis of CROA exists [4], it addresses only backscattering, without microscopic treatment. Meanwhile, microscopic studies [1–3] have focused primarily on specific models. Therefore, a unified microscopic framework covering all magnetic point groups and general scattering geometries is still lacking.

In this study, we first develop a microscopic formulation of CROA in localized spin systems. Decomposing the Raman operator into irreducible components for all 122 magnetic point groups, we develop general methods to determine whether CROA is allowed for given propagation directions of the incident and scattered light. We also show how such group-theoretical analysis helps identify the hopping processes and parameters that most strongly affect the magnitude of CROA. Finally, we apply this framework to real materials to perform a quantitative analysis of CROA.

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