Dichroic f-Sum Rule and the Orbital Magnetization of Many-Electron Systems

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Magnetic circular dichroism (MCD), the difference in absorption of left- and right-circularly-polarized light by magnetic systems, is described by $\sigma''_{\mathrm{A},\alpha\beta}(\omega)$, the imaginary, antisymmetric part of the optical conductivity. We obtain a sum rule for the MCD spectrum of many-electron systems which is the dichroic counterpart of the f-sum rule for linearly polarized light:

$$\int_0^\infty \boldsymbol{\sigma}''_{\mathrm{A}}(\omega)d\omega = \frac{\pi ec}{\hbar}\mathbf{M}_{\mathrm{CD}},$$

where $\sigma''_{\mathrm{A},\gamma} = (1/2)\epsilon_{\alpha\beta\gamma}\sigma''_{\mathrm{A},\alpha\beta}$. The conventional wisdom is that the total ground state orbital magnetization $\mathbf{M}_{\mathrm{orb}}$ should appear on the right-hand-side [1]. Instead, we find that the sum rule yields a subtly different quantity $\mathbf{M}_{\mathrm{CD}}$, which turns out to be one of the two contributions to $\mathbf{M}_{\mathrm{orb}}$ recently identified in the theory of bulk magnetism [2–4]:

$$\mathbf{M}_{\mathrm{orb}} = \mathbf{M}_{\mathrm{CD}} + \boldsymbol{\Delta}\mathbf{M}.$$

$\boldsymbol{\Delta}\mathbf{M}$ is generally nonzero for systems with more than one electron, and $\mathbf{M}_{\mathrm{CD}}$ is basically the wavepacket self-rotation term of Ref. [2]. By independently measuring their sum via gyromagnetic experiments, $\mathbf{M}_{\mathrm{CD}}$ and $\boldsymbol{\Delta}\mathbf{M}$ can in principle be separated out in macroscopic samples with a net magnetization. The dichroic f-sum rule is discussed alongside three other sum rules for optical absorption, including one which relates the dichroic spectrum to the interband (Karplus-Luttinger) Hall conductivity in solids. The sum rules for bounded and extended systems are first considered separately. We then show that for conventional insulators the bulk sum rules can be recovered from the bounded-system ones by taking the thermodynamic limit using a Wannier-function representation.

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