On field dependence of nonreciprocity in optical spectra of CuB_2O_4

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Energy levels and wave functions

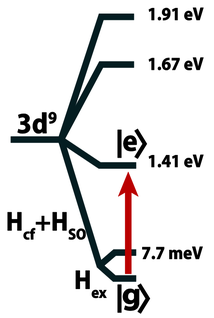

Fig. 1. Scheme of energy splitting of Cu^{2+} (^2D) [1,2].

$$\hat{H} = \sum_{k,q} B_q^{(k)} \hat{C}_q^{(k)} + \lambda \hat{\boldsymbol{l}}\hat{\boldsymbol{s}} + J_{gg}\hat{P}_g \sum_{i=1}^{4} \langle \boldsymbol{s_i} \rangle \hat{\boldsymbol{s}} + \mu_B(\hat{\boldsymbol{l}} + 2\hat{\boldsymbol{s}})\boldsymbol{B}$$

Crystal field parameters [2]: $B_0^{(2)}$ = −2.20 eV, $B_0^{(4)}$ = 1.23 eV, $B_4^{(4)}$ = 1.74 eV, the spin-orbit interaction parameter λ = −12 meV, exchange integral J_{gg} = 3.62 meV, $\hat{P}_g$ is the projection operator onto the ground orbital state. Wave functions for crystal field and spin-orbit interactions:

$$|g\rangle = 0.997 \cdot |\varepsilon_+\rangle + 0.069i \cdot |\zeta_+\rangle + 0.025 \cdot |\eta_-\rangle - 0.025i \cdot |\xi_-\rangle$$

$$|e_\pm\rangle = 0.985 \cdot |\zeta_\pm\rangle + 0.112i \cdot |\eta_\mp\rangle \pm 0.112 \cdot |\xi_\mp\rangle \pm 0.062i \cdot |\xi_\pm\rangle$$

Interaction with light

Broken time-inversion symmetry:

$$\hat{m} = \mu_B(\hat{\boldsymbol{l}} + 2\hat{\boldsymbol{s}})\sqrt{\epsilon\mu}[k \times E^\omega]$$

Broken space-inversion symmetry:

$$\hat{d} = \sum_{k,p,t} \{E^{(1)}\hat{U}^{(k)}\}_t^{(p)} D_t^{(1k)p}$$

Refractive index for magnetoelectric media depends on wave vector direction [3] ($E^\omega \parallel e_\alpha$, $B^\omega \parallel e_\beta$):

$$n_{\alpha\beta}^{\pm k} \approx \sqrt{\epsilon_{\alpha\alpha}\mu_{\beta\beta}} \pm \frac{1}{2}\left[\chi_{\beta\alpha}^{me} + \chi_{\alpha\beta}^{em}\right]$$

$$\chi_{\beta\alpha}^{me} \sim \langle g|\hat{m}_\beta|e\rangle\langle e|\hat{d}_\alpha|g\rangle \neq 0$$

Nonreciprocity in absorption spectra

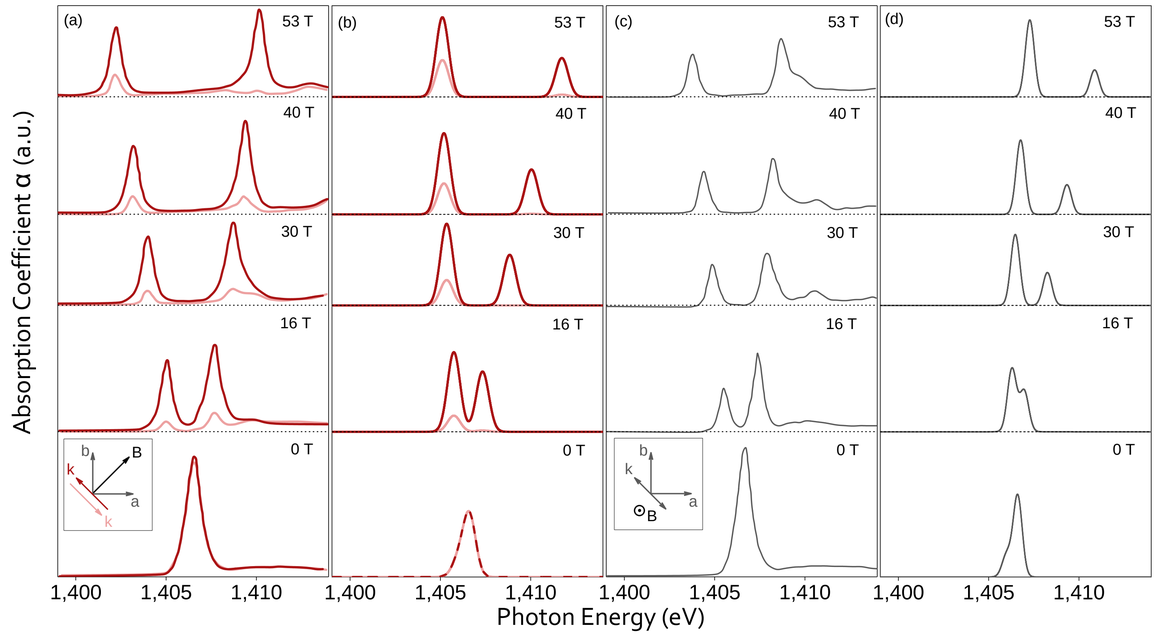

Fig. 2. (a, c) absorption spectra of CuB2O4 measured in [4], experimental setup is shown in the insets; (b, d) corresponding results of our calculations without taking into account the unclear origin of the bias in the high-frequency part of the spectrum. Theoretical lines shift toward higher frequencies as in experiments [5].

Directional asymmetry of luminescence

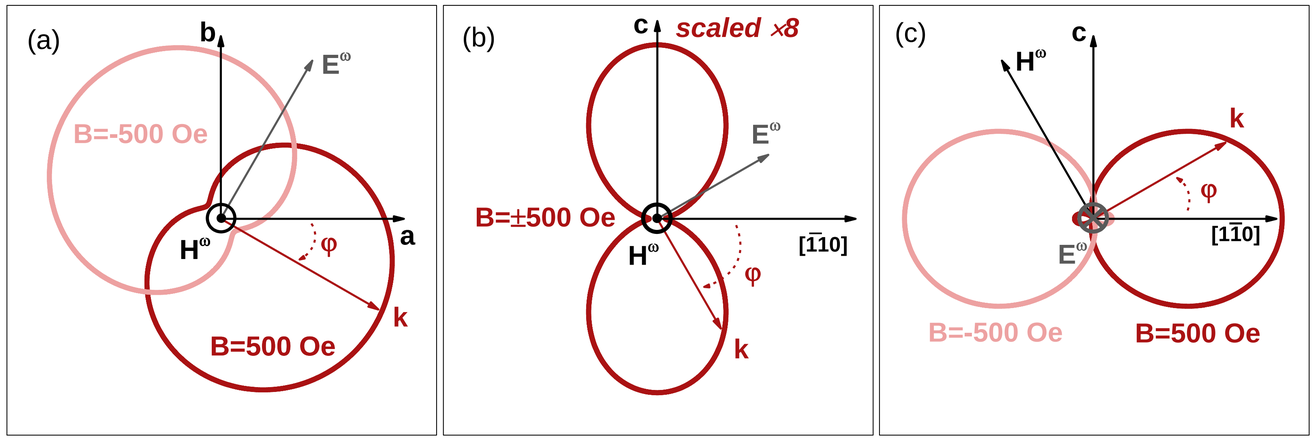

Fig. 3. Calculated luminescence diagrams of CuB2O4 for 1.41 eV transition. (a) is in good agreement with experimental data [6]

Results

A microscopic theory of dynamic magnetoelectric coupling in CuB2O4 is proposed. The energy levels, the $Cu^{2+}(3d^9)$ wave functions in an antiferromagnetically ordered sublattice of copper ions, and the probabilities of magnetic and electric dipole transitions are calculated, and the invariant components of the magnetoelectric coupling tensor are determined for different directions of the magnetic field. Photoluminescence intensity diagrams are calculated for different directions of the wave vector of light.

References

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