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Spectra and relaxation of electronic excitations in CsCdBr₃:Yb³⁺ and CsCdBr₃:Nd³⁺ monocrystals

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Abstract

Experimental and theoretical studies of the optical and EPR spectra, and the spin-lattice relaxation in CsCdBr₃ crystals containing 0.1, 0.3 and 1.0 mol.% of impurity Yb³⁺ ions have been fulfilled. In the optical excitation spectrum, a broad charge transfer band has been detected in the wave-number range of 25000-35000 cm⁻¹. The hybridization of the excited electron configuration corresponding to the charge transfer from the ligand (Br⁻) 4p closed shells on the ground 4f¹³ configuration of the ytterbium ion is considered to interpret the anomalously large crystal field splitting of the 2F_{5/2} multiplet. The EPR spectra with comparable intensities of the axial single-ion and symmetric dimer centres in the sample containing 0.3 mol.% of impurity Yb³⁺ ions were observed. Spin-lattice relaxation rates and linewidths in the EPR spectra of the single-ion and dimer centres were measured in the temperature range of 1.5 - 35 K at the frequency of 9.5 GHz at different directions of the applied magnetic field relative to the crystal symmetry axis. Experimental results are analyzed in the framework of the microscopic theory of the electron-phonon interaction with taking into account peculiarities of the phonon spectrum of the impurity CsCdBr₃ lattice. High-frequency EPR spectra of dimer centres in CsCdBr₃ crystals containing 0.2 and 0.5 mol.% of impurity Nd³⁺ ions were taken in the range of 205-250 GHz at 4.2 K in the magnetic fields up to 0.8 T parallel to the crystal symmetry axis. The crystal field splitting between the first excited and the ground Kramers doublets, and magnetic splitting factors of these doublets were determined. An estimate of the isotropic ferromagnetic exchange constant $A = (2.3 \pm 0.3) \cdot 10^{-3} \text{ cm}^{-1}$ in symmetric dimer centres formed by impurity Nd³⁺ ions was obtained from the zero-field splitting of the EPR signals.

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Keywords

Charge transfer, Crystal field energy, CsCdBr₃:Nd³⁺, CsCdBr₃:Yb³⁺, EPR, Isotropic exchange