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Photoluminescent and optical properties of ZnSe crystals doped with Yb and Gd rare-earth elements (REEs) and Cr impurity are investigated in infrared (IR) spectral range. The influence of stoichiometric deviation on photoluminescence (PL) spectra of the crystals is investigated and the structure of complex IR PL bands is analysed. The good coincidence between the IR PL spectra of the samples doped with Yb, Gd, and Cr is shown. Correlation between the component parts of the bands at 1 and 2 µm is found and possibility to control the composition of IR PL spectra by enrichment of the samples with Zn or Se is discussed. The model that explains the formation of associative centres based on the REEs and background Cu impurity fixed in the nodes of crystal lattice with tetrahedral symmetry, which are responsible for IR PL bands, is proposed. (© 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim)