LiYF$_4$:Ho crystal in a magnetic field: high resolution optical spectroscopy

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Abstract. Low-temperature high-resolution optical spectra of LiYF$_4$:Ho in an external magnetic field $H$ are presented. A field-dependent transformation of the hyperfine structure of holmium spectral lines is studied for the cases $H\parallel c$ and $H\perp c$. We show that Zeeman and hyperfine interactions lead to a strong coupling between crystal-field levels.

The spectra of rare-earth-doped LiYF$_4$ crystals demonstrate uniquely narrow lines, which gives possibility to detect very fine effects. In particular, for LiYF$_4$:Ho crystals, observation and study of the hyperfine structure due to interactions between the $4f$ electrons of the Ho$^{3+}$ ion and nuclear moments of the holmium nucleus [1-3] and isotope effects [4] have been reported. As far as we know, the spectra of LiYF$_4$:Ho subjected to an external magnetic field were not yet studied. In this work, we have performed a high-spectral-resolution optical study of LiYF$_4$:Ho in an external magnetic field. Isotopically enriched $^7$Li$_{0.16}$Li$_{0.9}$YF$_4$:Ho (0.1 at.%) single-crystal samples were investigated in permanent magnetic fields $H\parallel c$ and $H\perp c$ ($H = 0.6$ and 0.9 T).

Figure 1 shows the spectral line corresponding to the allowed electric dipole $\Gamma_{34} ({}^5I_8) \rightarrow \Gamma_2 ({}^2I_7)$ optical transition of the Ho$^{3+}$ ion. Clearly visible additional structure within hyperfine components results from an isotopic disorder within the lithium sublattice [4]. Each of the eight hyperfine components is doubly degenerate in a zero magnetic field [1,2]. A magnetic field splits the $\Gamma_{34}$ ground state causing a splitting of the spectral line. Interestingly, the intensities of the high-frequency hyperfine components are lower than those of the low-frequency ones, though the Boltzman distribution of populations suggests an inverse situation. The reason for this is that the splitting of the ground state diminishes the energy interval to the first excited state $\Gamma_2$ and, thus, enhances an admixture of its wave functions to the wave functions of the ground state. The $\Gamma_{34} ({}^5I_8) \rightarrow \Gamma_2 ({}^2I_7)$ transition borrows its intensity from the strong $\Gamma_{34} ({}^5I_8) \rightarrow \Gamma_3 ({}^2I_7)$ transition, the more the nearer is a given hyperfine sublevel of the ground level to the $\Gamma_2 ({}^2I_7)$ level.

An interaction between crystal-field levels, which is enhanced in a magnetic field, manifests itself also in unequal hyperfine intervals in the high- and low-frequency parts of the split line (see Figs. 1 and 2) and in appearance and/or growth in intensity of forbidden lines with $\Delta m = \pm 1$ (here, $m$ is a projection of the nuclear spin).

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Fig. 1. The $\pi$-polarized spectra in the region of the $\Gamma_{34}(^5I_8) \rightarrow \Gamma_{3}^{(2)}(^5I_6)$ transition of LiYF$_4$:Ho$^{3+}$ at 3.5 K in zero magnetic field (dashed trace) and in a magnetic field $H\|c$, $H = 0.6$ T (solid trace). Relative intensities of the hyperfine components simulate a violation of the Boltzman distribution.

Fig. 2. The $\pi$-polarized spectra in the region of the $\Gamma_{34}(^5I_8) \rightarrow \Gamma_{3}^{(2)}(^5I_6)$ transition of LiYF$_4$:Ho$^{3+}$ at 3.5 K in zero magnetic field (dashed trace) and in a magnetic field $H\|c$, $H = 0.6$ T (solid trace), demonstrating a dependence of the hyperfine intervals on the magnetic field. Inset displays a part of the spectrum in the magnetic field $H = 0.9$ T.

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References