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High-resolution spectroscopy of $^7LiYF_4$:Ho in a magnetic field: the first optical observation of the hyperfine levels' anticrossing

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The levels' anticrossing (avoided levels' crossing) takes place in different systems (atomic and molecular gases, semiconductors, molecular and atomic impurity centers in insulators) when a certain continuous parameter in the Hamiltonian (e.g., the magnetic field) varies. It is caused by a weak interaction, which can be considered as a perturbation, and manifests itself by the appearance of gaps in the spectrum of excitations. Anticrossing of levels causes many interesting phenomena. In particular, in solid state physics, the anticrossing of electron-nuclear (hyperfine) levels leads to pronounced effects in the experiments on optical orientation in semiconductors, to macroscopic tunneling of magnetization in rare-earth doped crystals. Until now, as far as we know, the anticrossing of hyperfine levels has been explicitly observed only in the EPR spectra. In this presentation, we report the first direct observation of hyperfine levels' anticrossing in the optical spectrum.

The effect was observed in the spectrum of the $^7LiYF_4:Ho^{3+}$ single crystal in an external magnetic field. Lithium-yttrium fluorides doped with rare-earth ions are widely used in various fields of photonics, and they are also unique model systems for studying various phenomena and interactions. It should be noted that these crystals are characterized by a record-low inhomogeneous broadening of spectral lines, which allows one to observe very subtle effects by optical methods. In particular, using the high-resolution Fourier spectroscopy for $^7Li_{1-x}{}^6Li_xYF_4:Ho^{3+}$ crystals, the effect of isotopic disorder in the matrix on the spectra of the monoisotopic Ho^{3+} ion was investigated.

We measured low-temperature (5 K) absorption spectra (in the energy range of 5000–15000 cm^{-1}, with a resolution of up to 0.001 cm^{-1}) of the $^7LiYF_4:Ho^{3+}$ (0.1 at.%) monoisotopic crystal in a magnetic field (up to 180 mT) parallel to the tetragonal crystal symmetry axis [1,2]. At the points of the expected crossing of the electron-nuclear levels differing in the projections of the nuclear spin by $\Delta m = 2$ and $\Delta m = 0$, gaps of 0.01 - 0.06 cm^{-1} (300 MHz - 1.8 GHz) were observed. We carried out a simulation of the spectra based on the Hamiltonian, which included the energy of a free ion, and crystal-field interaction, Zeeman, electron-deformation, hyperfine magnetic dipole and electric quadrupole interactions. It was shown that the anticrossings with $\Delta m = 2$ are due to the $A_J(J_xI_x + J_yI_y)$ part in the hyperfine interaction, and information was obtained on the hyperfine structure of the electronic singlet and on the electric quadrupole interaction. Anticrossing at $\Delta m = 0$ are of a different nature – they are caused by random deformations of the crystal lattice and carry information about the distribution function of deformations in the crystal. This can be used for a sensitive control of the quality of crystals, which is necessary, for example, for applications in quantum information science. Anticrossing of hyperfine levels at $\Delta m = 2$ provide the basis for constructing Λ and V three-level systems for optical quantum memory with equal probabilities of transitions in the arms.

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References:

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