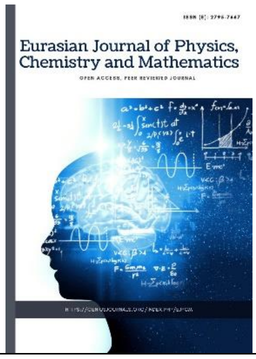

Features Of Optical And Magneto-Optical Spectra Of Rare-Earth Ions Ho^{3+} And Tb^{3+} In Garnet Crystals

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ABSTRACT

The article is devoted to the analysis of the results of experimental and theoretical studies of the relationship between magneto-optical and optical-magnetic effects with the features of the Stark structure of the energy spectra of non-Kramers RE ions in garnet crystals.

Keywords: magneto-optics, rare earth ion, Stark sublevels, Faraday effect, absorption, radiation, 4f-4f transition, spectrum.

Numerous studies have established that the optical and magneto-optical properties of RE paramagnetic garnets for sufficiently low excitation energies (<40,000 cm-1) are due to forbidden 4f-4f transitions between the Stark sublevels of the ground 4f(n) configuration, the degeneracy of which is partially (or completely) removed by the crystal field (CF) of the environment [1]. Further removal of the degeneracy from the Stark sublevels of the ion and "mixing" of their states by an external magnetic field H leads, on the one hand, to the emergence of circular anisotropy of the optical absorption spectra and the associated Faraday effects (FE), magnetic circular dichroism (MCD), and on the other hand, to circular anisotropy of the emission spectra - magnetic circular polarization of luminescence (MCPL). Analysis of the results of the MCD and MCPL methods, in the case where the width of the absorption (or emission) line significantly exceeds the value of the Zeeman splitting, makes it possible to identify optical transitions, to obtain important information about the nature of the energy levels between which the optical transition occurs, the symmetry of their wave functions, and the spectroscopic parameters of the radiative states of RE ions in crystals.

The wide possibilities of magneto-optical methods prove to be extremely useful and informative in the detailed study of energy spectra and symmetry of electronic states of non-Kramers RE ions in crystals. This is due to the fact that the application of modern methods of optical spectroscopy of RE compounds encounters certain difficulties in interpreting the optical spectra of RE ions with an even number of 4f electrons, since the experimentally observed number of components in their spectra and the intensity distribution in them do not coincide with theoretical predictions. Often the reason for this is the occurrence in the energy spectra of non-Kramers ions, the so-called quasidoublets formed by two closely located Stark singlets, which are not directly resolved in optical experiments.

To a large extent, the difficulties in interpreting the optical spectra of non-Kramers ions in garnets can be overcome if the forbidden 4f-4f transitions are studied by methods of linear differential magneto-optical spectroscopy: MCD, MCPL, etc. On the one hand, the main source of MCD and MCPL is the "mixing" by an external magnetic field H of the wave functions of closely spaced non-degenerate Stark sublevels of the ground and excited multiplets of a non-Kramers ion, combining in an optical transition, which allows positioning the "quasi-doublet" states in the energy spectrum of a RE ion. On the other hand, the application of symmetry selection rules for matrix elements of optical transitions provides a unique opportunity to determine the symmetry of the wave functions of the Stark sublevels from which (or to which) optical transitions occur [2]. Therefore, the complex use of optical and magneto-optical research methods allows us to obtain important experimental information, practically inaccessible to other physical research methods, about excited electronic states of non-Kramers RE ions in garnet crystals; to substantially deepen and clarify existing ideas about the relationship of magneto-optical effects with the Stark structure of the energy spectra of non-Kramers RE ions in garnet crystals and the physical mechanisms of record magneto-optical and optical-magnetic effects in non-Kramers RE ions.

The relationships between magneto-optical and optical-magnetic effects and the features of the Stark structure of the energy spectra of non-Kramers RE ions in garnet crystals have been studied experimentally and theoretically. In this regard, the following problems have been solved [2-4]:

- Experimental study of the spectra of the degree of MCPL on radiative 4f-4f transitions^{5}D4→7F6And^{5}D4→7F5in terbium – yttrium aluminum garnet Tb0.2Y2.8Al5O12 at temperature T=90 K.
- Study of the influence of the "Ising" character of magnetization of Stark sublevels of some quasi-doublet states of multiplets^{5}D4 and 7F5the main 4f(8)-electron configuration of the non-Kramers RE ion Tb3+ in the CP of D2 symmetry on the occurrence of a significant degree of circular anisotropy of the secondary glow of the paramagnetic garnet Tb0.2Y2.8Al5O12 in a magnetic field at low temperatures.
- Experimental study of optical absorption and luminescence spectra of terbium gallium garnet Tb3Ga5O12 in the visible and near ultraviolet (UV) spectral regions at temperatures T=78 K and 300 K in order to clarify the Stark structure and symmetry of the wave function of excited quintet states of multiplets^{5}D4, 5D3, 5G5, 5G6, 5L9 and 5L10 main4f(8)-electron configuration of the rare earth non-Kramers ion Tb in the garnet-gallate matrix. The optical resolution of the measuring setup used was no worse than ~1.5÷2 cm-1 in the wavelength ranges studied.
- Conducting numerical calculations of energy spectra of multiplets^{7}FJ (J=6.5, ... 1.0), 5D3, 5D4, 5G6, 5G5, 5L9 and 5L10 main4f(8) -electron configuration of the Tb3+ rare earth ion in the crystal field of D2 symmetry and interpretation on their basis of the features of the optical absorption and luminescence spectra of the terbium gallium garnet Tb3Ga5O12 in the visible and near ultraviolet (UV) spectral regions.
- Experimental study of the possible influence of an external magnetic field on the oscillator strengths of radiative 4f-4f transitions^{5}D4 →7F5And^{2}S2 →5I8in crystals of YAG:Tb3+ and YAG:Ho3+ garnets with a quasi-doublet structure of the energy spectrum of non-Kramers RE ions.

The forbidden (by parity) radiative 4f-4f transitions, on which the MCPL phenomenon is observed in the YAG:Tb3+ garnet, obey the electric dipole selection rules and that the dominant contribution to the mechanism of “unbanning” (by parity) of magneto-optically active radiative 4f-4f transitions in the YAG:Tb3+ garnet is made by “admixture” of the odd-symmetric component of the CP symmetry D2 of the states of the mixed excited 4f(7)5d(l′=2) configuration to the states of the ground 4f(8)(l=3) configuration of the Tb3+ RE ion.

"Ising" character of the behavior of Stark sublevels of some quasi-doublet states of multiplets^{5}D4 and 7F6 main4f(8) – electron configuration of non-Kramers RE ion Tb3+ in the CP of D2 symmetry promotes the occurrence of a significant degree of circular anisotropy of secondary luminescence of paramagnetic garnet Tb0.2Y2.8Al5O12 in a magnetic field. A similar correlation between the "Ising" character of Kramers doublets of RE ion Dy3+ in the garnet structure and significant values of the degree of MCPL, recently discovered in aluminate Dy0.2Y2.8Al5O12, indicates a certain degree of universality and applicability of this empirical regularity in magnetooptics of RE compounds.

The influence of an external magnetic field on the oscillator strengths of 4f→4f transitions in YAG:Tb3+ and YAG:Ho3+ garnet crystals is a fairly common phenomenon in RE systems with a quasi-doublet structure of the energy spectrum. As a result, the contribution of "mixing", which is usually neglected when considering magneto-optical effects on the absorption lines of RE ions, near the luminescence lines of Tb3+ and Ho3+ ions can play a significant role in the magnetooptics of YAG:Tb3+ and YAG:Ho3+ garnets, leading to anomalously large, by the standards of radiative magnetooptics, values of the circular anisotropy of the secondary luminescence spectra in a magnetic field.

The results of temperature studies of the optical density spectra D and luminescence, compared with the data of numerical calculations of the energy spectra of excited states of the RE ion Tb3+ in a crystal field of D2 symmetry, made it possible to establish the Stark structure and the nature of the symmetry of the wave function of excited quintet states of multiplets^{5}D3, 5D4, 5G6, 5G5, 5L9 and 5L10 main4f(8) – electron configuration of the non-Kramers RE ion Tb3+ in the yttrium gallate garnet matrix.

The scientific value of the results is determined by the fact that they make it possible to explicitly reveal the relationship between the features of the Stark splitting of the energy spectra of non-Kramers RE ions Tb3+ and Ho3+ and the physical mechanisms of record magneto-optical and optical-magnetic effects in non-Kramers RE ions in YAG:Tb3+ and YAG:Ho3+ garnet crystals.

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