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Scintillation properties of LuAG-based scintillators: Influence of Ga-admixture, non-stoichiometry and Mg-codoping

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Scintillators based on lutetium-aluminum garnet LuAG:Ce have been studied in past decades. Besides many favorable properties, slow decay components related to retrapping of electrons on the shallow traps related to anti-site defects in caused deterioration of scintillation parameters. These defects, where Lu resides at Al sites, are created at high temperatures during crystal growth from the melt. One of the strategies of improvement was Ga and Gd admixture leading to highly-efficient GAGG scintillators. Ga admixture lowered the edge of the conduction band burying the trapping levels. Gd admixture increased the separation of the Ce³⁺ 5d1 level from the conduction band decreasing the probability of 5d-state ionization. Another strategy was Mg-codoping, which stabilized the tetravalent Ce⁴⁺, which can directly compete for electrons in the early stage of scintillation mechanism, suppressing the slow components and increasing the light yield. Another way to improve the scintillation properties of LuAG:Ce can be the shift of the crystal stoichiometry towards the Al-rich compositions. There is also a question if combination of such stoichiometry and Mg codoping or Ga admixture can bring further improvement.
Ce, Mg-codoped samples grown from Al-rich melt (1at% and 3at% excess) were prepared by micro-pulling-down method to study the influence of non-stoichiometry. Ga admixed LuGAG crystals with the same Ce and Mg concentrations were grown as well together with the Al-rich analogs.
Codoping of Mg at a concentration around 300 ppm led to improvement of overall scintillation efficiency, which is attributed to the alternative scintillation mechanism involving Ce⁴⁺ stabilized by Mg²⁺.
Non stoichiometry 3 at% in the LuAG matrix for the Ce0.2% Mg 300 ppm sample led to significant improvement of the overall scintillation efficiency (almost a factor of 2) when compared to the stoichiometric samples. Exceptionally high light yield (17000 photons/MeV) under 667 MeV gamma-ray excitation (6 μs amplifier shaping time) was observed when compared to the standard stoichiometric LuAG:Ce crystals (5000 photons/MeV). Lower light yield values are due to lower quality crystals grown by the micro-pulling-down method. As was shown by the measurement of the scintillation decay under pulsed soft X-ray excitation, the highest Mg concentration brings additional slow components to the scintillation decay and did not bring any other improvement in general.
Very similar trends were observed for the Ga-admixed LuGAG, but the anti-site defect-related luminescence was strongly suppressed, which led to further improvement of the overall scintillation efficiency.
Thermally-stimulated luminescence (TSL) and temperature dependences of photoluminescence characteristics were measured for selected samples. Interestingly, tunneling of electrons towards Ce luminescence center from the adjacent defects was observed after excitation to the lowest Ce³⁺ 5d state. For the non-stoichiometric sample, decrease of the steady-state luminescence intensity was observed at low temperatures around 150K, while for the stoichiometric sample the quenching starts above 450K. On the other hand, the photoluminescence decay time starts to decrease above 600K for all the samples, which points to the fact that a non-relaxed excited state of Ce³⁺ can be involved. Luminescence and scintillation mechanism in relation with composition and stoichiometry will be presented and discussed.

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