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Non-radiative relaxation of rare-earth ions through coupling to hydrogen and deuterium impurities in crystals

Thulium-doped lithium niobate is a leading candidate for implementation of quantum information and signal processing systems. The wide absorption profile and exceptionally large oscillator strength are ideal for high-bandwidth and frequency-multiplexed applications. In many of these applications, optical waveguides allow chip-scale integration of optical elements. A popular method for fabricating waveguides in LiNbO3 is to diffuse hydrogen into the crystal; however, hydrogen can affect the optical properties of other ions, such as thulium, leading to unwanted non-radiative relaxation and heating effects. To determine the effect of hydrogen impurities on thulium-doped LiNbO3 crystals, we in-diffused thulium ions into the surface of LiNbO3 wafers and studied the fluorescence of the 3H4 - 3H6 Tm3+ transition for varying concentrations of hydrogen. Controlled amounts of hydrogen were added to the LiNbO3 wafers using proton exchange methods and the change in the thulium fluorescence lifetime was measured using pulsed laser excitation. Results suggest a very strong short-range interaction between thulium and hydrogen, causing rapid non-radiative relaxation for thulium near hydrogen and a reduction in lifetime by orders of magnitude. In contrast, there is no observable effect on thulium that is further from the hydrogen. Consequently, while increasing hydrogen levels reduces the fraction of thulium with long lifetimes needed for quantum and classical signal processing, the lifetimes of the remaining unaffected ions are still suitable for these applications. Results suggest that the fabrication of waveguides in thulium-doped LiNbO3 by proton exchange methods may be a viable approach for quantum and classical information applications over some ranges of hydrogen concentrations.

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