Power dependence of upconversion luminescence in lanthanide and transition-metal-ion systems

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We show theoretically with the simplest possible model that the intensity of an upconversion luminescence that is excited by the sequential absorption of n photons has a dependence on absorbed pump power P, which may range from the limit of P^n down to the limit of P^1 for the upper state and less than P^1 for the intermediate states. The two limits are identified as the cases of infinitely small and infinitely large upconversion rates, respectively. In the latter case, the dependence of luminescence intensities from intermediate excited states on pump power changes with the underlying upconversion and decay mechanisms. In certain situations, energy-transfer upconversion and excited-state absorption can be distinguished by the measured slopes. The competition between linear decay and upconversion in the individual excitation steps of sequential upconversion can be analyzed. The influence of nonuniform distributions of absorbed pump power or of a subset of ions participating in energy-transfer upconversion is investigated. These results are of importance for the interpretation of excitation mechanisms of luminescent and laser materials. We verify our theoretical results by experimental examples of multiphoton-excited luminescence in $Cs_3Lu_2Cl_9:Er^{3+}$, $Ba_2YCl_7:Er^{3+}$, $LiYF_4:Nd^{3+}$, and $Cs_2ZrCl_6:Re^{4+}$.